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ANDC ANALYSIS IN BEAM-FOIL SPECTROSCOPY

by

© ROBERT NELSON GOSSELIN

A THESIS

SUBMITTED TO THE FACULTY OF GRADUATE STUDIES AND RESEARCH

IN PARTIAL FULFILMENT OF THE REQUIREMENTS FOR THE DEGREE

OF Master of Science

IN

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THE UNIVERSITY OF ALBERTA

FACULTY OF GRADUATE STUDIES AND RESEARCH

The undersigned certify that they have read, and recommend to the Faculty of Graduate Studies and Research, for acceptance, a thesis entitled ANDC ANALYSIS IN BEAM-FOIL SPECTROSCOPY

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in partial fulfilment of the requirements for the degree of Master of Science in Physics.

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ABSTRACT

The problem of determining atomic lifetimes from beam-foil decay curves which are subject to cascading effects is discussed. The ANDC method of analysis, which in theory can exactly account for all cascade effects, is described and a practical method of applying it is developed. Several quantities which result from the analysis, and the possible variations on the method of analysis, can be used as self-consistency checks on the applicability of the method to a given situation.

The results of a test on three decay situations using synthesized data are given along with examples of its application to real data. Using these results, several rules for applying the ANDC method and interpreting the results are developed. Recommendations for obtaining experimental decay curves most suited for ANDC analysis are also made. The computer program developed to perform the analysis is included along with instructions for its use.

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My thanks to Dr. G.L. Cumming for making available the two dimensional fitting routine (alias CORREG), which provided a means of including error correlation effects in the analysis.

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CHAPTER I

INTRODUCTION

An important complement to any theory is its verification by experiment. The application of quantum mechanics to many-electron atoms is not a straightforward task. Many assumptions and approximations are necessary in order to calculate atomic energy levels.[1] Before the advent of beam-foil spectroscopy[2], hereafter referred to as BFS, the amount of experimental data available for comparison was relatively small due to the limitations of the conventional sources (shock tubes, electric discharges and furnaces). The main advantages of BFS are that one can, in principle, excite almost any ionic state of any element and that one can also follow the decay of that state with time. This enables one to study trends in oscillator strengths along various sequences (e.g., isoelectronic and homologous). Comparisons of these experimentally determined trends with theoretical predictions provide a good test of the accuracy of the approximations used in the quantum mechanical calculations (CCF 76). Accurate oscillator strengths for neutral and highly ionized atoms are also needed for the determination of astrophysical quantities (e.g., particle

---

[1] Theorists work in terms of transition probabilities A_{ij} or oscillator strengths f_{ji}, while BFS experimentalists determine lifetimes. The relationships between the three quantities are given in section 1.1 below.

[2] Pioneered by S. Bashkin in 1964 (Ba 64). See also the recent review article on BFS (Be 77).

densities) and for studies in laser and plasma physics.

The past ten years have seen four international conferences devoted entirely to BFS.[1] Major topics at these conferences have been the determination of atomic lifetimes, and possible methods to improve the accuracy of these determinations. It should be mentioned that BFS has also been used to determine fine and hyperfine structures, Lande g-values and for general spectral analysis of ionized atoms. Examples of such studies may be found in these conference reports.

1.1 The Beam-Foil Technique

A mono-energetic beam of ions, having velocity typically in the range from 1 to 5 mm/ns, is magnetically analyzed for the desired mass and directed through a thin foil (usually of carbon and having a thickness of typically 500A). The interaction with the foil results in a beam of ions at various levels of excitation and ionization, depending on the beam energy. The excited states then decay by the emission of radiation and the intensity of a particular transition can be measured as a function of time (see Fig. 1.1).

The basic data recorded is a series of triplets (d_i, N_i, T_i), where d_i is the distance from the foil and N_i is the number of photons accumulated in time T_i. Time is

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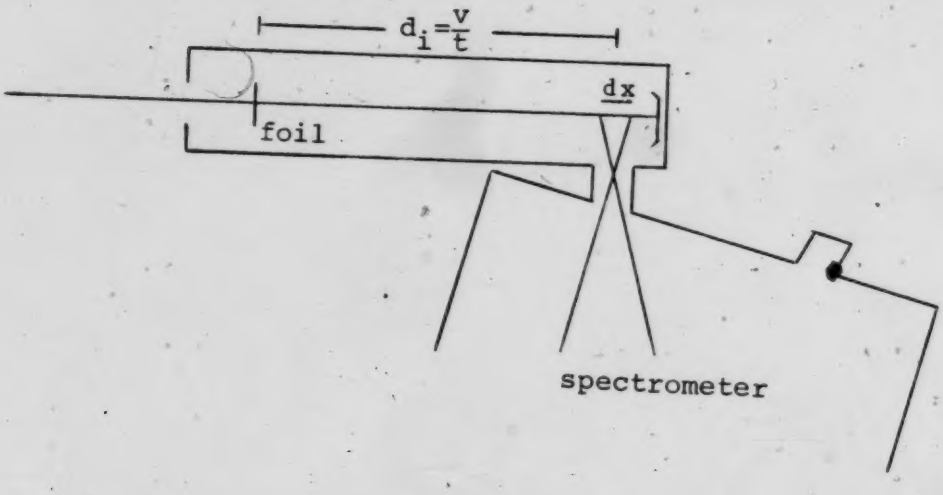
[1] Proceedings: Ba 68, MBB 70, Ba 73, SP 76.

Fig. 1.1 Basic beam-foil experimental configuration (not to scale).



determined by a pre-set level for integrated beam current as measured by the Faraday cup, i.e., intensity is normalized by beam current.

In the past, this photon counting procedure[1] has been the most common method of collecting beam-foil data. However, some experimental groups, including the one at the University of Alberta, are now using multi-channel analysis techniques to obtain more reliable data with greater efficiency. In the multi-channel analysis (MCA) method several sweeps (varying distance from the foil) are summed together to obtain the final decay curve. The main

---

[1] The general experimental design and procedures that have been used at the University of Alberta are described in IL 76.

advantage of this method is that during a run any significant change in the decay curve of the latest sweep, as compared to previous sweep(s), indicates a change in the excitation process (e.g., beam instability, change in foil condition) and the run can be terminated at this point without a significant loss of data. Thus the MCA method allows for the constant monitoring of the quality of the data and minimizes the time and foils spent in acquiring the data.

Obviously a primary condition for determining accurate lifetimes is the acquisition of the best decay data possible. However, the means of analyzing this data once it has been obtained is a separate problem and is the topic of this work. Thus details of experimental equipment and procedures will not be discussed.

If only one atomic level was excited, one could observe the exponential decay in intensity and thus measure the lifetime, τ, of that level directly from the slope of the logarithmic decay, since

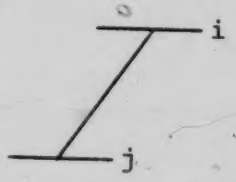
$$I_{ij}(t) \propto -dN_i/dt \quad \text{and}$$

$$N_i(t) = N_0 e^{-t/\tau} \quad \text{or} \quad \ln(I_{ij}) = -t/\tau + \text{const.}$$

where N_0 is a constant corresponding to the count which would be recorded at the foil position. In the event that the transition $i \to j$ is the only (or dominant) mode of decay

from level i, then τ_i is simply $1/A_{ij}$, where A_{ij} is the transition probability.

Fig. 1.2 Simple two level system.



This is related to the oscillator strength, f_{ji}, by the relation

$$A_{ij} = \frac{6.67 \times 10^{15}}{\lambda^2(A)} \frac{g_j}{g_i} f_{ji} \quad ,$$

where g_i, g_j are the statistical weights of the two levels. If more than one decay mode can significantly depopulate the level i, then $\tau^{-1} = \sum_j A_{ij}$. Conversion from τ_i to f_{ji} then requires a knowledge of the relative values of the A_{ij}, which can be obtained from branching ratio measurements or from theoretical calculations.

1.2 Problems of the Method

The wide range of states available with BFS is somewhat of a mixed blessing because of the non-selective nature of the excitation process. Levels above the one under consideration are also excited and decay into the level of

interest, repopulating it and masking the simple exponential behaviour of a single decay. This problem of cascading levels is the major difficulty in determining lifetimes. Common methods of dealing with the problem are described briefly in the next section.

Occasionally, two different transitions have similar wavelengths so that the lines overlap. This problem always occurs to some degree, but in most cases is not a significant source of error.

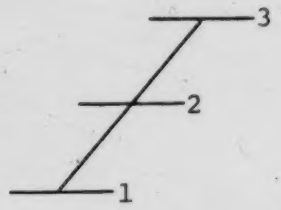
The beam density is low ($\sim 10^{+5} cm^{-3}$) so that trapped radiation, stimulated emission and ionic recombination effects are negligible (these are usual problems with many conventional sources). However, this means that the intensity is correspondingly low. Also, the foil ages and eventually breaks due to the action of the beam, giving a finite time in which one can collect data. This limits BFS to relatively strong transitions unless multi-channel analysis techniques are used (PWVL 77). The time scale is determined by the velocity of the beam, thus the accuracy of the lifetime is limited by the uncertainty in the velocity. Various techniques can be used to obtain the beam velocity to within 1%, which is usually less than the statistical error resulting from the cascading problem to be discussed in the next section.

1.3 Analysis of the Cascade Problem

In order to take into account the presence of

cascading, one must first examine its contribution to the decay curve. The simplest case which shows the effect of cascading is the three level system shown in Fig. 1.3 . The

Fig. 1.3 Three level decay scheme.



intensity of the primary decay is $I_{21}(t)=\alpha_2 N_2(t)$ but,

$$\frac{dN_2(t)}{dt} = -\alpha_2 N_2(t) + \alpha_3 N_3(0) e^{-\alpha_3 t} \quad ,$$

therefore

$$\frac{d(N_2(t)e^{\alpha_2 t})}{dt} = \alpha_3 N_3(0) e^{(\alpha_2-\alpha_3)t} \quad .$$

Integrating both sides of the equation with respect to time gives:

$$N_2(t)e^{\alpha_2 t} = \frac{\alpha_3}{\alpha_2-\alpha_3} N_3(0) e^{(\alpha_2-\alpha_3)t} + \text{const.}$$

Setting t=0, one finds that

$$\text{const} = N_2(0) - \frac{\alpha_3}{\alpha_2-\alpha_3} N_3(0) \quad .$$

Substitution of this gives the population of the primary level:

$$N_2(t) = \left[N_2(0) + \frac{\alpha_3}{\alpha_3-\alpha_2} N_3(0) \right] e^{-\alpha_2 t} + \frac{\alpha_3}{\alpha_3-\alpha_2} N_3(0) e^{-\alpha_3 t} \quad .$$

Thus the primary intensity is given by the expression:

$$\frac{I_{21}(t)}{\alpha_2} = \left[N_2(0) + \frac{\alpha_3}{\alpha_3-\alpha_2} N_3(0) \right] e^{-\alpha_2 t} + \frac{\alpha_3}{\alpha_3-\alpha_2} N_3(0) e^{-\alpha_3 t} \quad .$$

This shows a particular instance of the general problem: the decay curve has as many exponential terms as levels which cascade into it plus the one due to itself.[1] Usually a few of these terms dominate; however, this still leaves some difficulty in obtaining the desired lifetime.

The most obvious thing to do is to try to fit the decay curve to a function consisting of a sum of exponential terms. Usually it is not worthwhile or necessary to try more than three exponentials plus a constant background. This has its problems, however, since the fit is not unique, and one must make a careful analysis to find the "correct" parameters. Computer programs have been written to do this

---

[1] The general result for an arbitrary decay scheme is given in Appendix I.

analysis (e.g., HOMER see IL 74) and has been found to be adequate in most cases. The estimated errors in the parameters obtained from such fitting procedures are only valid if the form of the fitting function is correct (PDKI 78), and determining the correct form can be difficult when severe cascading is present.

Integral and differential techniques are useful when the primary lifetime is much shorter or longer than any of the significant cascades. If one differentiates the intensity curve, the relative amplitude of the term with the shortest lifetime is increased. Similarly, if the intensity curve is integrated the longest lifetime term increases in relative amplitude. This may be seen by the following example of an intensity curve with two exponential terms corresponding to lifetimes of two and ten seconds.

$$\text{e.g.} \quad I(t) = e^{-t/2} + e^{-t/10}$$

$$-\frac{dI(t)}{dt} = 0.5e^{-t/2} + 0.1e^{-t/10}$$

$$\int_0^t I(t')dt' = 12 - 2e^{-t/2} - 10e^{-t/10}$$

These curves can then be analyzed by simply looking at the regions where simple exponential behaviour is evident or by using the multi-exponential fitting method. Although these techniques appear promising at first sight, in practice they do not give more reliable values than the multi-exponential curve fitting method discussed previously. This is because

of the problems associated with the numerical differentiation of statistical data.

Another possible technique is to measure the decay of alignment of an excited state (BCS 72). This method assumes that transfer of alignment by cascades is small. A similar method, which is applicable even if cascade alignment does occur, uses the decay of g-value quantum beats (LC 72). Both these methods use only 1-10% of the available intensity and are of limited applicability. Finally, a coincidence counting experiment (MS 73) has also been tried, but the counting rate was found to be much too low for accurate lifetime determination.

A potentially useful technique was suggested by L.J. Curtis and his colleagues in 1970 (CBB 70, CBB 71). In this technique the decay curves for the primary transition and all its direct cascades are recorded and then analyzed simultaneously. In principle, this method can completely account for the effects of cascading on the value which is determined for the lifetime of the primary level. This method of analysis is the topic of the rest of this work.

CHAPTER II

THE ANDC METHOD

The application of the ANDC[1] theory to be derived in section 2.1 will, in principle, solve the problem of cascade effects in radiative lifetime measurements. However, the presence of experimental errors (i.e., statistical fluctuations of photon counts) and the finite amount of data means that there will be some error involved in the determination of the lifetime. Thus the problem is not only how to apply ANDC analysis to best advantage, but also to provide reliable error limits on the lifetimes obtained. Section 2.2 describes the calculation of the basic ANDC variables and associated errors, and also provides an introductory discussion of the relative merits of the different methods of calculating these variables.

2.1 ANDC THEORY

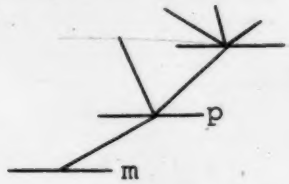
Consider the generalized decay scheme in Fig. 2.1, where a primary level (p) decays into a state (m) and the primary level has a number of higher levels decaying into it. These higher levels are also subject to cascading. By including all direct transitions (n→p) into the primary level, the population rate of change of the primary level is found to be

---

[1] Acronym for Arbitrarily Normalized Decay Curves. The reason for this name will be made apparent in section 2.1.

$$\frac{dN_p(t)}{dt} = \sum_n N_n(t) A_{np} - N_p(t)/\tau_p \quad . \qquad (2.1)$$

Fig. 2.1 Generalized Decay Scheme.



Noting that the observed intensity for the transition i→j is $I_{ij}=E_{ij}N_i(t)A_{ij}$, where E_{ij} is the relative detection efficiency[1] of the experimental apparatus for the transition, the following expressions can be substituted into equation 2.1 :

$$N_p(t) = \frac{I_{pm}(t)}{E_{pm}A_{pm}}, \quad N_n(t) = \frac{I_{np}(t)}{E_{np}A_{np}} \text{ and } \frac{dN_p(t)}{dt} = \frac{dI_{pm}/dt}{E_{pm}A_{pm}} \quad .$$

This gives the result

$$\frac{dI_{pm}/dt}{E_{pm}A_{pm}} = \sum_n \frac{I_{np}(t)}{E_{np}} - \frac{I_{pm}}{\tau_p E_{pm}A_{pm}} \quad ,$$

which can be written as:

---

[1] The detection efficiency is determined by the optical properties of the experimental apparatus used and depends on the wavelength of the transition.

$$\tau_p \frac{dI_{pm}}{dt} = \sum_n \xi_n I_{np} - I_{pm}, \text{ where } \xi_n = \frac{E_{pm} A_{pm}}{E_{np}} . \qquad (2.2)$$

Integration of equation 2.2 with respect to time between initial time, t_i, and final time, t_f yields:

$$\tau_p \{I_{pm}(t_f) - I_{pm}(t_i)\} = -\int_i^f I_{pm} dt + \sum_n \xi_n \int_i^f I_{np} dt \ ;$$

$$\text{or} \quad P/\Delta = \sum \xi_n C_n/\Delta + \tau_p \ , \qquad (2.3)$$

where $P \equiv \int I_{pm}(t)dt$ = area under primary curve,

$C \equiv \int I_{np}(t)dt$ = area under cascade curve

and $\Delta \equiv I_{pm}(t_i) - I_{pm}(t_f)$ = change in primary intensity

This last equation is linear in the variables P/Δ and C/Δ, whose values depend on the choice of t_i and t_f (see Fig. 2.2), with constant coefficients 1 and ξ_n respectively[1].

The constant term is the primary lifetime, which can now be obtained by performing a linear fit using the variables P/Δ and C/Δ. If no errors were present in the data, a simple linear fitting procedure could be used. However, as mentioned previously, the presence of experimental errors requires the use of special fitting procedures to account

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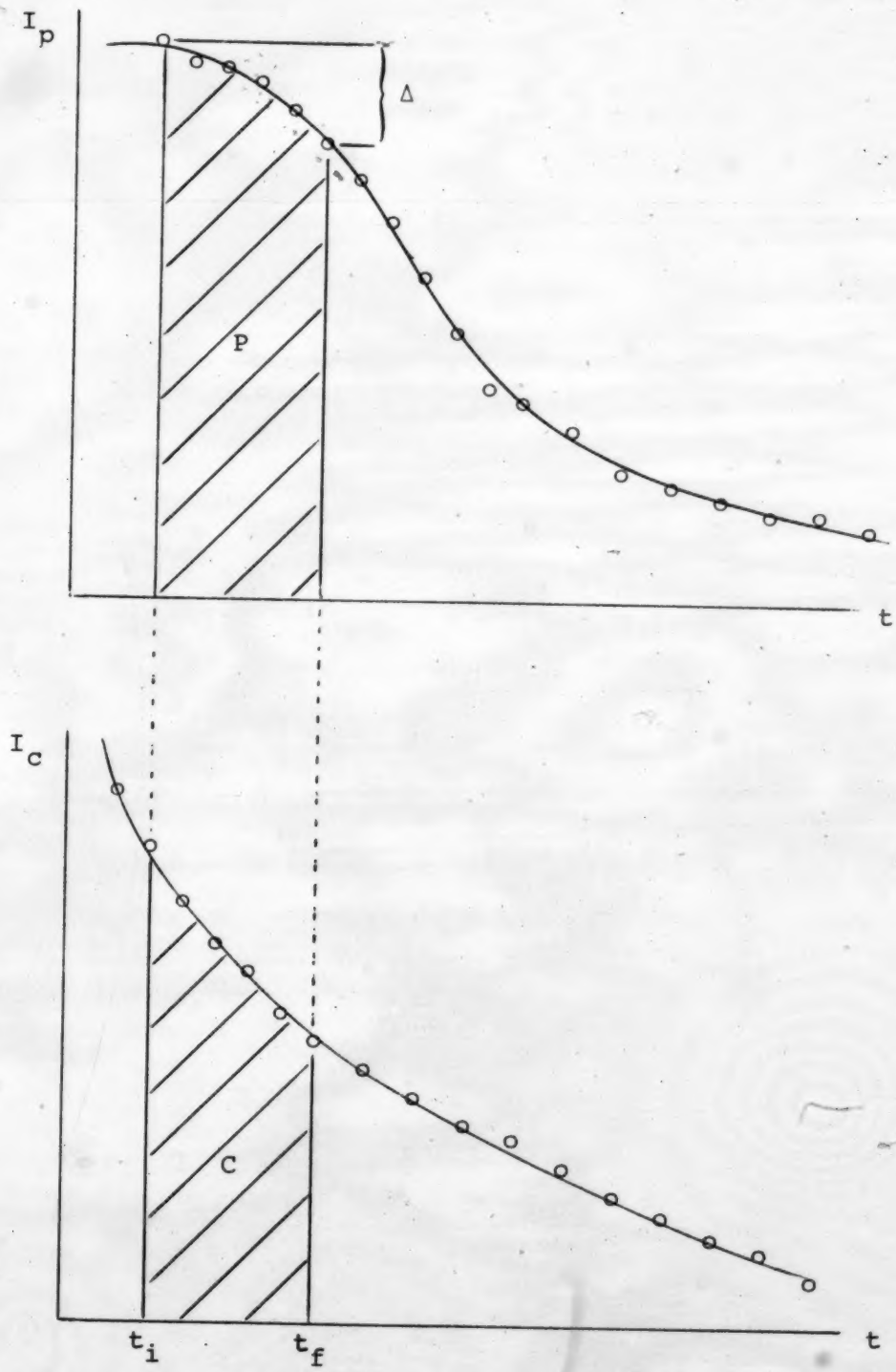
[1] Note that since the ξ_n contain the relative detection efficiencies of the transitions, the absolute normalization of the decay curves is irrelevent in determining the lifetime.

Fig. 2.2 Example of an ANDC panel.



properly for these errors.

It is possible to write equation 2.3 in the form

$$\Delta = \frac{P}{\tau} - \sum_n \frac{\xi_n}{\tau_p} C_n \quad . \tag{2.4}$$

This equation is not as simple as 2.3, since it has an additional variable term. However, it does have certain advantages due to error considerations, as is shown in the next section.

2.2 Practical Application of ANDC

The basic quantities (P,C,Δ), which are the areas under the decay curves and the change in the primary intensity, are most easily calculated directly from the signal data. To do this, the cascade and primary intensities must be measured at the same distances from the foil. If this is not the case, either interpolation between data points or a smooth fit to the cascade decay curve can be used to calculate the cascade intensity at distances determined by the primary data.[1] Linear interpolation between distance coordinates is preferable to fitting the curve for the reasons given below and in Chapter III.

The principal objection to using a fitted curve,

---

[1] The ANDC program for the case of one direct cascade presented later in this work allows the six parameters of a HOMER multi-exponential fit to be used to "reconstruct" the cascade data set.

particularly for the primary, is that any subsequent error calculation in the ANDC analysis may be of doubtful validity. The reason for this is that the estimated errors in the fit parameters usually reflect how good the fit is as a whole, even though the fit may be poor (or excellent) in a particular portion of the decay curve. In other words, since the true decay curve is a complicated sum of exponential terms, which is difficult to fit even without statistical fluctuations, any condensation of the information into a few parameters will generally lead to a loss of information about particular parts of the curve. This loss can be particularly important when calculating the intensity difference, Δ.

The areas are calculated using the trapezoidal rule:

$$P = \sum_{j=i}^{f-1} \frac{(Y_j+Y_{j+1})(t_{j+1}-t_j)}{2} \quad , \text{ where } Y \equiv \text{intensity.}$$

Thus the variance[1] in the area is given by

$$P = \sum_{j=i}^{f-1} \frac{(Y_j+Y_{j+1})(t_{j+1}-t_j)^2}{4} \quad ,$$

since there is no correlation between the intensities at different times and $V(Y_j)=Y_j$ due to Poisson statistics.

The difference, $\Delta_{if}=Y_i-Y_f$, has a very large relative error when compared to that of P_{if} or C_{if}, since $V(\Delta)=Y_i+Y_f$.

---

[1] Notation: variance $V(X)=\langle(dX)^2\rangle$.

For example, if Y_i=9000 and Y_f=8000,

$$S(P)/P = (9000+8000)^{\frac{1}{2}}/17{,}000 = .008 \text{ , whereas}$$

$$S(\Delta)/\Delta = (9000+8000)^{\frac{1}{2}}/(9000-8000) = .130 .$$

This is especially true when Δ is small. To minimize this error, the intervals t_i-t_f(referred to as panel sizes) are made as large as possible and a single exponential fit is performed upon the five point section of the decay curve centered on the desired point in order to obtain Y_i or Y_f.

This fit is necessary in the tail of the decay curve, since it can happen that the measured Δ is less than zero due to the statistical fluctuations. The fit also reduces the error in Δ since five points are being used to obtain the intensity at one point. Thus we can divide the variance by the number of degrees of freedom (5-2=3).

There are two possible ways to proceed from this point. Considering the simplest case of one direct cascade and referring to equations 2.3 and 2.4, one can either fit points of the form (Δ,P,C) OR (P/Δ,C/Δ). The latter choice looks simpler in that it requires only a two dimensional linear fit, whereas the other requires a three-dimensional fit. However, dividing both areas by Δ not only introduces large error to both variables, but also causes almost 100%

correlation between the errors of the two variables.[1]

Assuming a linear relationship between X and Y, it is possible to write an expression for the probability of obtaining the set of data points (X,Y) and associated errors (<dX>,<dY>,<dXdY>). This probability will be a function of two parameters, the slope and the intercept of the line. Maximization of this probability with respect to the two parameters will yield equations for them which can be solved numerically. The errors in the two parameters can be estimated by a similar procedure (CRRW 72). This procedure results in a satisfactory lifetime and associated standard deviation in cases where it is applicable, that is, when there is only one significant direct cascade present. Examples of its application are given in subsequent chapters, where this is referred to as the (2-D) method of analysis.

In the case of only one direct cascade, one can eliminate much of the correlation between the variables of the two dimensional fit by writing the ANDC equation in the form

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[1] There is also a small correlation between P and Δ, but it has a negligible effect on the results.

$$Y = \tau^{-1} - \sum_n \frac{\xi_n}{\tau} X \text{ , with } Y = \Delta/P \text{ and } X = C/P \text{ .} \qquad (2.5)$$

This reduces the correlation of errors of X and Y and also reduces the error in X, since the largest source of error, Δ, is now only used to calculate Y and not X. However, tests on real data showed no significant difference the lifetimes and errors obtained using this equation[1] instead of the (2-D) method, when the method is applicable. This occured despite the fact that the correlation coefficient, $R=\langle(dX)(dY)\rangle/(S(X)S(Y))$, for each panel was reduced from about 0.95 to 0.1 in going from the (2-D) to the (2-A) method. This is a good indication that the error calculation and weighting procedure of the linear fitting program is valid, even when the correlation between the variables is very large. A test was done on a case which had two direct cascades, but the analysis was made including only one. Both methods of two dimensional analysis produced unacceptable (but different) results. The alternate two dimensional method may be of use when the error estimation procedure of fitting routine has trouble converging.

In the case of the three variables (Δ,P,C), there is no significant correlation among the variables. Since the errors in the areas are very small ($<<10\%$) compared to the

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[1] Referred to as the (2-A) method later on in this work.

error in Δ, the multi-linear fit can be weighted using only the variance of Δ.[1] The fitting procedure is more straightforward than the two-dimensional case above, but the errors cannot be accounted for rigorously. The obvious advantage to this procedure is that it can easily be extended to cases where more than one direct cascade is present.

There are also two distinct ways of partitioning the decay curve into several panels. The panels can be overlapping or non-overlapping. The latter choice severly restricts the size and number of panels (i.e., data points), but produces data points which are completely independent of each other. Overlapping panels will result in some correlation between data points, which will make the fitting procedure and error estimates obtained from it less rigorous.

The next two chapters will be devoted to testing these possible variations of the ANDC method on synthetic data (Chapter III) and actual experimental data (Chapter IV).

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[1] This is very convenient, as there is no statistically valid procedure for performing a multi-linear fit if the errors in the variables are comparable. The computer program used is REGRES ,which is taken from Bevington (Be 69).

CHAPTER III

ANDC ANALYISIS OF SYNTHETIC DATA

This chapter establishes the usefulness of the ANDC method of analysis in situations where significant cascading is present. Conventional methods of analysis may give poor or misleading results in these cases and ANDC can serve as a valuable check on the results.

In order to assess the accuracy and reliability of the different ways of applying the ANDC method described in the previous chapter, a few appropriate situations where the lifetimes are exactly known are necessary. This is most easily accomplished using artificially constructed decay curves. Comparisons of the ANDC results with the known values and the results obtained using conventional curve fitting techniques reveal that ANDC is an important tool in the analysis of cascaded situations.

3.1 Synthetic Data

The model decay schemes used are shown in Figs. 3.1-3.3. The initial populations of the levels were chosen to be proportional to $(2l+1)/n^3$, which previous studies indicate is approximately the case in many physical situations.[1] Branching ratios and lifetimes were chosen to represent some typical situations where cascading effects are important. The analytic expressions for the intensities

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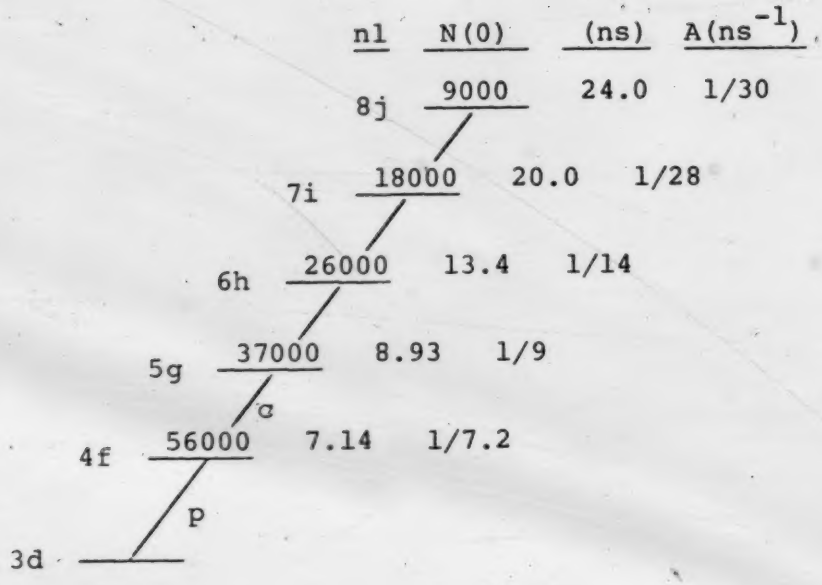
[1] (DMV76,LSW72,LC73,DDD70)

Fig. 3.1 Synthetic Data: case 1.



Primary lifetime=7.14ns
Primary branching ratio=0.993
Primary transition probability=0.139ns^{-1}

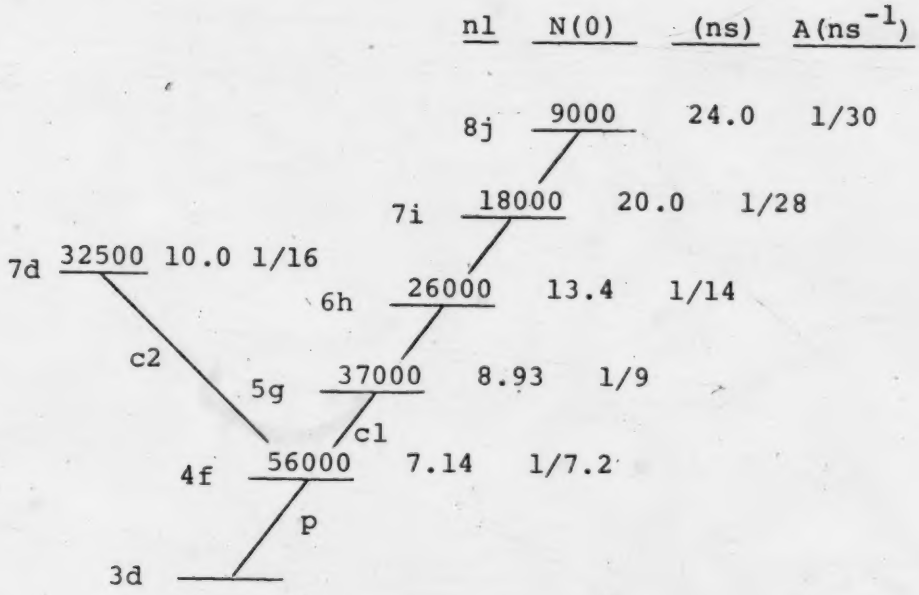
Primary intensity curve is defined by

$$I_p(t) = -516.5e^{-t/7.14} + 1328e^{-t/8.93} + 5906e^{-t/13.4} + -5155e^{-t/20} + 6215e^{-t/24}.$$

Cascade intensity curve is defined by

$$I_c(t) = 268.4e^{-t/8.93} + 2782e^{-t/13.4} - 3342e^{-t/20} + + 4403e^{-t/24}.$$

Fig. 3.2 Synthetic data: case 2.



Primary lifetime=7.14ns
Primary branching ratio=0.993
Primary transition probability=0.139ns^{-1}

Primary intensity curve is defined by

$$I_p(t) = -7560e^{-t/7.14} + 1328e^{-t/8.93} + 5906e^{-t/13.4} + -5155e^{-t/20} + 6215e^{-t/24} + 7043e^{-t/10}.$$

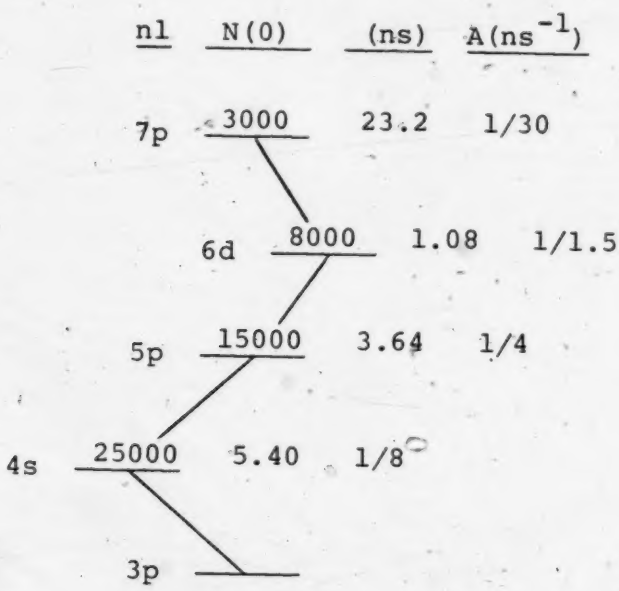
Cascade intensity curve(#1) is defined by

$$I_{c1}(t) = 268.4e^{-t/8.93} + 2782e^{-t/13.4} - 3342e^{-t/20} + + 4403e^{-t/24}.$$

Cascade intensity curve(#2) is defined by

$$I_{c2}(t) = 2031e^{-t/10}.$$

Fig. 3.3 Synthetic data: case 3.



Primary lifetime=5.40ns
Primary branching ratio=0.675
Primary transition probability=0.125ns^{-1}

Primary intensity curve is defined by

$$I_p(t) = 10652e^{-t/5.4} - 7939e^{-t/3.64} + 340e^{-t/1.08} + 71.71e^{-t/23.2}.$$

Cascade intensity curve is defined by

$$I_c(t) = 5687e^{-t/3.64} - 2018e^{-t/1.08} + 81.5e^{-t/23.2}.$$

were obtained from $N_i(0)$, A_{ij} and τ_i using the general formula given in Appendix I and assuming $I_{ij}=A_{ij}N_i(t)$.

Each data set has sixty points and all sets corresponding to the same case have the same set of distance coordinates. The program used in the analysis assumes that the data are subject to Poisson statistics, and it calculates errors and weights points accordingly. This fluctuation is present in all real data, so an attempt is made to make the synthetic data fluctuate about the "true" value according to Poisson statistics by using the FUZZ subroutine of the program HOMER (IL 74).

3.2 Analysis

The analysis was performed in several different ways in order to determine which would give the most accurate results. For the two cases (Figs. 3.1,3.3) where only one direct cascade was present, both the two-dimensional (2-D) and multi-dimensional (M-D) methods were used. The (2-D) method was not used for the case shown in Fig. 3.2, since the presence of a second direct cascade would render the results inconclusive.

The data points were partitioned into panels three different ways, with a varying degree of overlap between the panels. The number of ANDC points[1] was kept at about

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[1] The term "ANDC point" refers to the calculated quantities (P,C,Δ) associated with a particular panel (cf. Fig 2.2).

fourteen for each partitioning. Table 3.1 gives the three different partitionings used, (A,B,C), where the numbers define the endpoints of the panels and refer to the data points in order of increasing distance from the foil.

Table 3.1 Panel divisions for synthetic data analysis.

A paneling contains 15 ANDC points:
1 5 10 15 20 25 30 35 40 45 50 53 32 37 20
10 20 25 30 35 50 45 42 47 52 56 60 36 40 60

B paneling contains 14 ANDC points:
1 5 9 13 17 24 29 34 38 46 51 20 30 50
4 8 12 16 23 28 33 37 45 50 54 30 40 60

C paneling contains 14 ANDC points:
1 6 9 13 17 21 25 29 33 37 41 46 51 56
5 8 12 16 20 24 28 32 36 40 45 50 55 60

The first partitioning, A, has a large degree of overlap between successive panels, which allows for large panels each having 10-15 points. The second partitioning, B, has non-overlapping panels for all but the last three ANDC points, which overlap most of the preceeding panels. The last partitioning, C, provides no overlap at all, but consequently gives smaller panel sizes, typically having about 5 points per panel.

The program yields the primary lifetime in each case, plus quantities which would be the branching ratio in the (2-D) analysis or the transition probability in the (M-D) analysis, provided that the detection efficiencies for the

primary and cascade were the same (cf. eqns. 2.3,2.4) The estimated errors in these quantities are also obtained, along with the value of the reduced chi-squared for the fit.[1]

An important question now arises. How is it possible to decide when a particular analysis is unacceptable? An unacceptable fit may occur due to poor data or when the analysis performed is inconsistent with the actual physical situation, e.g., the analysis is performed assuming only one direct cascade when two are really present.

One indication of the validity of the analysis is the value of the reduced chi-squared (χ^2). The probability that the fit obtained is valid is small when the chi-squared is significantly greater than one.

In the multi-dimensional analysis, the fit allows for an "extra" constant term, K,

$$\Delta = K + \frac{P}{\tau} - \frac{1}{\tau}\sum_n \xi_n C_n$$

which should be equal to zero. If the value obtained for K differs significantly from zero (considering the estimated error in K and the typical value of Δ used in the analysis), then the analysis should be rejected. Also, the multi-dimensional analysis assumes that the error in the area is

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[1] See Appendix III for a more detailed description of the program output.

negligible compared with that in Δ. The validity of this assumption should also be checked in each analysis. In the two-dimensional analysis, the plot of P/Δ versus C/Δ should consist of points randomly scattered about a straight line. The presence of a blend or additional direct cascade will generally result in a curved rather than a straight line plot, thus giving another criterion for rejection of an analysis. The sign of the transition probability (modified by efficiency factors) should obviously be positive, giving a further check on the validity of the fit.

3.3 ANDC Results: Synthetic Data

The results of the analyses are shown in Tables 3.2-3.4 and reveal several important features. Most of the values obtained for the lifetimes, transition probabilities and branching ratios fall well within the estimated single standard deviations and all values fall within two standard deviations of the true value. It is apparent from Tables 3.2-3.4 that the standard deviation is dependent on the average panel size; i.e., the larger the typical panel size, the smaller the standard deviation. The results also indicate that the estimated standard deviations on the lifetime values are consistent with both the deviations of the individual values from the true result and the scatter among the four values obtained for each individual partitioning pattern (A,B,or C) used. The degree of overlap given by the A partitioning appears to be close to the

Table 3.2 Synthetic data case 1: ANDC lifetimes.

SYN#1 (τ=7.14, A=.14, BR=.99)

PERM	τ	σ	A	δA	χ^2	
AA	6.90	0.36	0.15	0.01	0.97	(M-D)
AB	6.71	0.36	0.16	0.01	1.00	A PANELS
BA	7.16	0.38	0.14	0.01	1.37	extensive
BB	7.14	0.39	0.14	0.01	1.99	overlap
AA	7.61	0.68	0.13	0.02	0.71	(M-D)
AB	7.65	0.69	0.13	0.02	0.88	B PANELS
BA	7.17	0.67	0.14	0.02	0.87	MODERATE
BB	7.29	0.70	0.14	0.02	1.22	overlap
AA	7.12	1.01	0.14	0.03	0.53	(M-D)
AB	7.43	1.05	0.13	0.03	0.62	C PANELS
BA	7.13	1.02	0.14	0.03	0.74	no
BB	7.72	1.16	0.13	0.03	1.07	overlap
			BR	δBR		
AA	6.68	0.52	1.04	0.05	0.76	
AB	6.64	0.53	1.06	0.05	0.82	(2-D)
BA	6.78	0.50	1.03	0.05	1.01	A PANELS
BB	6.63	0.52	1.05	0.05	1.29	
AA	7.29	0.73	0.98	0.07	0.79	
AB	7.29	0.75	0.98	0.07	0.83	(2-D)
BA	6.76	0.72	0.92	0.07	0.63	B PANELS
BB	6.79	0.74	0.94	0.08	0.84	
AA	6.49	0.94	1.04	0.09	0.54	
AB	6.43	0.94	1.06	0.09	0.59	(2-D)
BA	6.66	1.09	1.03	0.10	0.44	C PANELS
BB	6.72	1.19	1.03	0.11	0.63	

optimum, giving the lowest estimated standard deviations with reasonable values for the reduced chi-squared; i.e., the panels are sufficiently large to give small errors, while maintaining a reasonable degree of independence between adjacent panels.

Another important feature is the pattern followed by

Table 3.3 Synthetic data case 2: ANDC lifetimes.

SYN#2 (τ=7.14, A=.14, BR=.99)

PERM	τ	σ	A	δA	χ^2	
AAA	7.03	0.53	0.15	0.02	1.1	
AAB	6.87	0.52	0.15	0.02	.95	
ABA	6.76	0.53	0.15	0.02	1.1	
ABB	6.70	0.52	0.15	0.02	1.1	(M-D)
BAA	7.80	0.61	0.12	0.02	.63	A PANELS
BAB	7.65	0.60	0.13	0.02	.52	
BBB	7.45	0.60	0.13	0.02	.57	
BBA	7.50	0.60	0.13	0.02	.58	
AAA	9.16	3.30	0.08	0.06	.43	
	.	.	.	.	.	(M-D)
	.	.	.	.	.	B PANELS
	.	.	.	.	.	
REJECTED DUE TO LARGE ERRORS AND LARGE CONSTANT TERM						
AAA	7.25	0.96	0.14	0.03	.63	
AAB	7.16	0.96	0.14	0.03	.68	
ABA	6.78	0.95	0.15	0.03	.70	
ABB	6.75	0.96	0.15	0.03	.78	(M-D)
BAA	8.59	1.41	0.10	0.03	.38	C PANELS
BAB	8.49	1.40	0.10	0.03	.38	
BBB	8.20	1.50	0.11	0.03	.47	
BBA	8.23	1.48	0.11	0.04	.49	

the permutations of primaries and cascades. The value of the lifetime obtained depends, in most cases, mainly on the primary and only slightly on the cascade matched with it. This is not unexpected since the primary contributes two variables (P and Δ) whereas a particular cascade affects the result through only one variable (C). Furthermore, the major source of uncertainty lies in , which is derived from the primary decay only.

In order to obtain the best estimate of the lifetime,

Table 3.4 Synthetic data case 3: ANDC lifetimes.

SYN#3 (τ=5.40, A=.125, BR=.675)

PERM	τ	σ	δA	δA	χ^2	
AA	5.40	0.11	.124	.006	1.8	
AB	5.43	0.11	.124	.006	1.8	(M-D)
BA	5.44	0.12	.122	.007	0.6	A PANELS
BB	5.46	0.12	.122	.007	0.5	
AA	5.15	0.23	.139	.012	.71	
AB	5.21	0.23	.139	.012	.73	(M-D)
BA	5.46	0.23	.119	.011	.23	B PANELS
BB	5.51	0.23	.118	.011	.23	
AA	5.13	0.31	.125	.018	.93	
AB	5.14	0.31	.125	.018	.96	(M-D)
BA	5.42	0.33	.119	.018	.20	C PANELS
BB	5.42	0.33	.119	.019	.20	
			BR	BR		
AA	5.39	0.11	.659	.022	1.3	
AB	5.43	0.11	.660	.024	1.4	(2-D)
BA	5.43	0.11	.658	.024	.45	A PANELS
BB	5.45	0.11	.663	.025	.42	
AA	5.31	DNC	.690	DNC	.70	
AB	5.32	DNC	.697	DNC	.72	(2-D)
BA	5.50	0.17	.639	.040	.18	B PANELS
BB	5.51	0.17	.645	.040	.19	
AA	5.43	0.29	.557	.102	.51	
AB	5.46	0.30	.550	.106	.59	(2-D)
BA	5.55	0.32	.594	.110	.18	C PANELS
BB	5.57	DNC	.594	DNC	.19	

DNC--> error analysis routine did not converge.

some method of averaging the results of the different analyses performed is required. Of the results obtained, only the B paneling on SYN#2 (Table 3.2) had to be rejected according to the criteria described above. These analyses were rejected due to the very large estimated standard deviations (~30%) and also the occurence of negative

transition probabilities. For the remaining analyses, the χ^2 of each result was included by multiplying the estimated standard deviation in the lifetime by χ^2 when $\chi^2>1$. Using these modified error estimates, the final weighted average, τ, and its estimated standard deviation, σ, were calculated using

$$\tau = \sum_i \frac{\tau_i}{\sigma_i^2} \qquad \sigma^2 = \frac{\sum_i \frac{(\tau-\tau_i)^2}{\sigma_i^2}}{\sum_i 1/\sigma_i^2}$$

The results for all three case studies were reduced using this method and the final values obtained are given in Table 3.5. The (2-D) and (M-D) methods were averaged

Table 3.5 Lifetimes of synthetic data: summary.

CASE	2-D	HOMER TROY	M-D	2-D M-D
#1 τ=7.14	6.80 ±.22 3%	8. ±2 25%	7.17 ±.20 3%	6.98 ±.29 4%
#2 τ=7.14	N.A.	8.41 ±.39 5%	7.22 ±.45 7%	N.A.
#3 τ=5.40	5.44 ±.04 1%	5.02 ±.16 3%	5.40 ±.10 2%	5.42 ±.08 2%

separately and together where applicable. The results of the conventional method of analysis are also included for

comparison. The programs HOMER and TROY[1] were used for the multi-exponential analysis. TROY is a program which fits up to six exponentials plus a constant term to a decay curve. Unlike HOMER, it allows any number of the thirteen possible parameters to be fixed. This can be useful for finding the primary lifetime when the cascade lifetimes are known. Using the cascade and the primary lifetimes as determined by HOMER and/or ANDC as initial estimates for the parameters of the multi-exponential fit, it is sometimes possible to confirm the ANDC lifetime estimate. This is done by holding any combination of these lifetimes fixed to obtain a fit consistent with all the "known" lifetimes.

Table 3.5 shows that the ANDC analysis provides quite satisfactory results, as regards both accuracy and precision, in all three cases. It is worth noting that, as one might expect, the percentage error in the lifetimes increases as the decay scheme becomes more involved.

The method of ANDC analysis described in this chapter requires that both the primary and the cascade data have the same set of distance coordinates at which the intensity is measured. Unfortunately, not all experimental data is in this form, so the original cascade data set may have to be modified. This can be done by either simple linear interpolation between points or by use of a multi-exponential fit to the cascade. It is possible for the

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[1] Written by R.L. Brooks at the University of Alberta.

latter choice to significantly reduce the reliability of the ANDC results for the reasons explained in Chapter II. This is demonstrated by the result of doing the previous ANDC analysis with fitted curves rather than the original cascade data. The result is shown in Table 3.6. The lifetime

Table 3.6 ANDC lifetimes obtained with original and fitted cascade data.

CASE	ORIGINAL	FITTED	
	6.90 .36	6.94 .37	
#1	6.71 .36	6.50 .37	A
7.14	7.16 .38	7.05 .37	PANELS
	7.14 .39	6.72 .37	
	7.12 1.0	6.52 .96	C
	7.43 1.1	6.69 .96	PANELS
	7.13 1.0	6.59 1.0	
	7.72 1.2	6.67 .97	
AVERAGE:	7.01±0.2 (3%)	6.78±0.1 (1.5%)	
	5.40 .11	5.43 .11	
#3	5.43 .11	5.47 .11	A
5.40	5.44 .12	5.47 .12	PANELS
	5.46 .12	5.49 .12	
	5.13 .31	5.15 .31	
	5.14 .31	5.09 .31	C
	5.42 .33	5.44 .33	PANELS
	5.42 .33	5.39 .33	
AVERAGE:	5.41±.07 (1.4%)	5.44±.08 (1.6%)	

obtained for Case 1 from the analysis using the fitted cascade data set is misleading in that the error estimate is

much too small. However, the analogous reslt for Case 3 is essentially identical to the result obtained using the original cascade decay data.

Linear interpolation is more reliable since it cannot produce any systematic deviation from the actual experimental data (provided that the data points are taken at reasonable intervals). The linear interpolation will only affect the exact manifestation of the Poisson statistics of the data by reducing the random fluctuations, i.e., the cascade decay curve will be partially smoothed in the process of interpolation.

3.4 Summary

On the basis of the preceding analyses of synthetic data, several rules can now be given for the application of the ANDC method to real data. Firstly, it is more important to maximize the quality of the data (counting statistics, number of runs etc.) for the primary decay rather than for the cascade decays, since the calculated lifetime depends principally on the primary data set. Also, where posssible, the data should be taken so that the distance coordinates are the same for the primary and the cascade(s). If this last condition cannot be fulfilled, then it is preferable to use linear interpolation rather than a multi-exponential fit for the cascade.

During the actual analysis, the paneling should be done in at least two different ways, one with no overlap and one

with moderate (~50%) overlap between adjacent panels, e.g., Table 3.1, C and A panels respectively. The results of both panelings should be consistent, even though the overlap analysis will probably give the better results. It is also useful to check the results by using both the (2-D) and (M-D) analyses (where possible).

The following quantities can indicate that a particular analysis should be rejected.

i) reduced χ^2 --reject analysis if too large,

ii) curvature of graph (2-D analysis only) --reject if significant curvature present,

iii) "validity"(M-D only) --reject if these quantities <1 for too many points in the analysis,

iv) constant K (M-D only) -- reject if too large and/or estimated error in K is too small (considering the typical value of Δ); e.g., if $\Delta \sim 200$ and $K = 80 \pm E$; one would definitely reject this if $E \lesssim 10$ and would possibly accept it (depending upon the other criteria above) if $E \gtrsim 40$.

v) sign of transition probability -- reject if negative.

It is important to bear in mind that these rejection criteria are not infallible. It is always possible for circumstances to combine to give a physical situation which yields an "acceptable" result from an analysis which does not correspond to the physical situation in the experiment. For example, a significant direct cascade may have been left out of the analysis, yet the analysis may still be "acceptable" according to the criteria given above, thus

yielding a misleading value for the lifetime. An important safeguard in this regard is a study of the energy level scheme and the cascade scheme relevant to each particular primary lifetime measurement to insure that no significant cascade transition has been overlooked. No amount of mathematical manoeuvering on the computer can compensate for an oversight made in the laboratory.

In Chapter IV, examples will be presented showing the application of these general validity criteria to ANDC analyses performed on actual beam-foil decay curve data.

CHAPTER IV

ANDC ANALYSIS OF REAL DATA

This chapter provides examples of the application of ANDC analysis as described in the previous chapter to some real data. The examples include cases which were intended to be used in conjunction with ANDC analysis in that the primary and cascade distance coordinates are matched, and also cases where linear interpolation and/or multi-exponential fits had to be used for the cascade(s). However, before these examples can be discussed, one must first consider the possible effects of the instrumental time averaging that occurs in the collection of actual data.

4.1 Time Averaging Effects

Experimentally, one does not measure the intensity at a particular distance, d_i, from the foil; rather one measures an average intensity over a segment of the beam centered on the point, d_i. Fig. 4.1 is a schematic of the experimental situation when taking measurements below 2000A and it shows that the averaging time, δ, depends on the mask size, m, slit size, s, and the beam velocity, v. Mask sizes in this laboratory usually range from 5-52mm, slit sizes from 100-800m, with typical velocities in the 1-2 mm/ns range and d_s/d_m 0.06. Note that, due to this averaging time, the first meaningful data point after the foil occurs at time $t=\delta/2$ (unless one is able to include the experimental window function in the data analysis, THB 77). This results in

Fig. 4.1 Optical configuration for wavelengths <2000Å.
(not to scale)

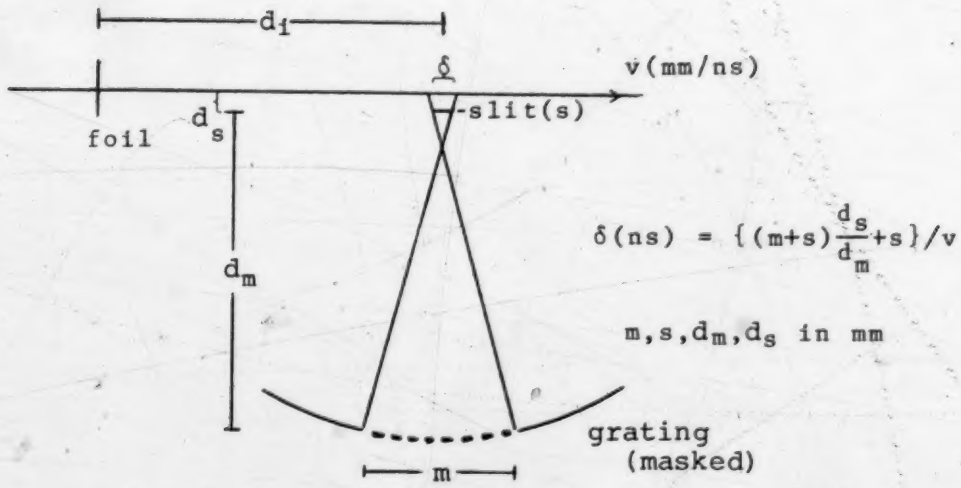
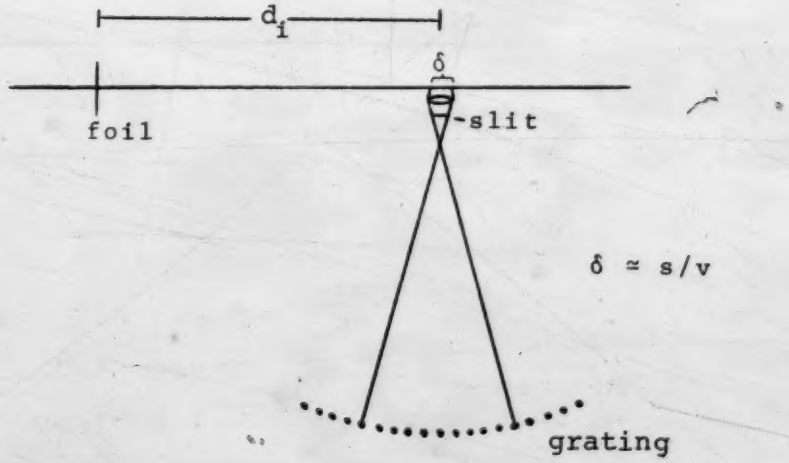


Fig. 4.2 Optical configuration for wavelengths >2000Å.
(not to scale)



values for δ ranging from about 0.2 to 4 ns. Fig. 4.2 shows the analogous situation when the wavelength is greater than 2000A. In this case $\delta \simeq s/v$ and $s<800$ m, so this case is included in the previous one.

Frequently, δ is made larger than is desirable, sacrificing good time resolution in order to obtain a sufficiently high counting rate. Thus when measuring lifetimes in the range less than 1 ns, δ is often as large as the value of the lifetime being measured. The question now arises as to whether this will affect the value obtained for the lifetime and, if so, what value must δ have relative to the lifetime so that the effect is significant?

As an example, take a two component decay curve with "ideal" intensity

$$I_i(t) = Ae^{-\alpha_1 t} + Be^{-\alpha_2 t} .$$

Integration over the segment, δ, yields the measured intensity

$$I(t) = A \frac{\mathrm{sh}(\alpha_1\delta/2)}{\alpha_1/2} e^{-\alpha_1\delta/2} e^{-\alpha_1 t} + B \frac{\mathrm{sh}(\alpha_2\delta/2)}{\alpha_2/2} e^{-\alpha_2\delta/2} e^{-\alpha_2 t} ,$$

where the factors of $e^{-\alpha_1\delta/2}$ and $e^{-\alpha_2\delta/2}$ are included so that the first valid data point corresponds to t=0. These two equations show that the shape of the decay curve will change depending on the values $\alpha_1\delta$ and $\alpha_2\delta$. Taking S, the ratio of the first and second coefficients, as a measure of

the shape of the curve,

$$S = \frac{A}{B} \frac{\mathrm{sh}(\alpha_1\delta/2)}{\mathrm{sh}(\alpha_2\delta/2)} \frac{e^{-\alpha_1\delta/2}}{e^{-\alpha_2\delta/2}} \frac{\alpha_2}{\alpha_1}$$

$$= \frac{A}{B} \frac{\alpha_2}{\alpha_1} \frac{1-e^{-\alpha_1\delta}}{1-e^{-\alpha_2\delta}}$$

The shape of the "ideal" curve is $S_i = A/B$.

A quantity which will reflect how the measured shape deviates from the ideal case is

$$R = \frac{S}{S_i} = \frac{\alpha_2}{\alpha_1} \frac{1-e^{-\alpha_1\delta}}{1-e^{-\alpha_2\delta}}$$

The value of R for some values of $\alpha_1\delta$ and $\alpha_2\delta$ are given in Table 4.1. It is apparent that in most cases the shape

Table 4.1 Values of the Shape Factor, R, for some $\alpha_i\delta$.

$\alpha_1\delta$	$\alpha_2\delta$	R
.05	.01	.98
.50	.01	.79
.50	.10	.87
.50	.30	.91
.75	.15	.76
1.0	.20	.70

of decay curves will change significantly ($R \lesssim .90$) if $\alpha_1\delta \gtrsim 0.5$. However, this will not invalidate the procedure of

multi-exponential fitting in order to obtain the lifetime since the various exponential terms are unchanged. But what happens to the ANDC analysis?

ANDC analysis states that, in the case of one direct cascade, the lifetime of the primary level is given by

$$\tau = \frac{P(I,F)}{\Delta_{IF}} - \xi \frac{C(I,F)}{\Delta_{IF}} .$$

If Δ is such that one or both decay curves change shape, then the quantities (P,C,and Δ) become (P',C', and Δ'). Is it then true that

$$\tau = \frac{P'(I,F)}{\Delta_{IF}} - \xi \frac{C'(I,F)}{\Delta_{IF}} ?$$

If not, how much deviation from the true value of the lifetime can be expected?

An analytic calculation was done which suprisingly enough shows that the value of δ has absolutely no effect on either the value of τ or ξ obtained from ANDC analysis. The details of this computation can be found in Appendix III.

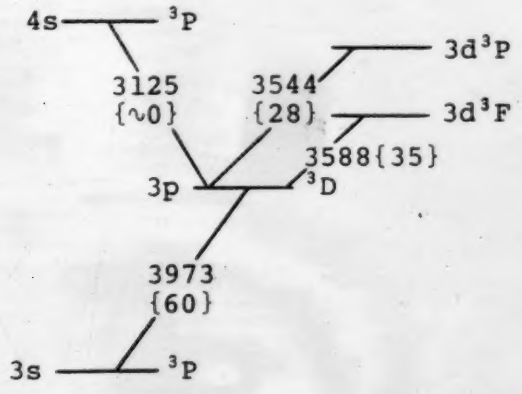
Of course, in practice one cannot make δ arbitrarily large for basically the same reason as one cannot do so when using a multi-exponential fit, the reason being that the information from the component with lifetimes much less than δ would be lost in the larger random fluctuations of the data, resulting in very large uncertainties in the values computed for them.

To summarize this section, one can conclude that there is no special difficulty in applying ANDC analysis to decay curves which have been significantly affected by time averaging. Of course, just as for multi-exponential curve fitting, the uncertainty in the primary lifetime will increase as the averaging time, δ, is increased. Furthermore, for the ANDC analysis to remain valid, it is necessary that the same averaging time be used for both the primary and the cascade(s) in a given ANDC data set.

4.2 Fluorine II[1]: $3p^3D$ Level

The relevant cascade decay scheme involving the $3p^2D$ level is shown in Fig. 4.3. The ANDC analysis was performed

Fig. 4.3 Decay scheme of F II - ^3D level (numbers in {} are approximate intensities).



[1] All the data used for the analyses in this chapter were obtained at the Radiation Research Laboratory of the University of Alberta.

using the transition (3973A) to the $3s^3P$ level as primary and the line at 3588A as the only direct cascade. The line at 3125A was not detectable, which indicates that cascading effects due to this transition were small. The direct cascade at 3544A was severly blended with the transition $3d^3D$-$5p^3P$ and $3s^3D$-$3p^3P$ and therefore could not be included in the ANDC analysis. It was, however, weak relative to the 3588A line and therefore should not be a major contributor to repopulation of the $3p^3D$ level.

Three runs were done on both the primary and the cascade, giving nine permutations of primary with cascade. The beam velocity after the foil was 2.08mm/ns (±2%) with the beam energy at about 1.0 MeV. There were between 40 to 48 data points in each of the primary data sets with peak counts of around 4400. The background count rate was similar for all three primaries (~4 per sec.), but the average collection time per point was a factor of two larger for primary #1 (P#1). The cascade data sets have 38 data points each and peak counts between 3000 and 4000. The last data point taken, for both primary and cascade, corresponds to a time since excitation of about 40 ns.

Two types of partitioning were used for the ANDC analysis, as indicated in Table 4.2, which also contains the results of the analysis. These data were not taken with ANDC in mind, so the cascade decays were replaced with fitted curves in the analysis. Both multi-exponential

Table 4.2 Results of ANDC Analysis on F II Data.

	(2-D) (MHF)			(2-D) (L-I)			(M-D) (L-I)			
PC	τ	σ	χ^2	τ	σ	χ^2	τ	σ	χ^2	
11	6.14	.17	6.1R	6.21	.16	3.9R	7.42	.29	1.3X	
12	6.13	.16	3.8R	6.14	.16	3.6R	7.35	.31	1.4X	
13	6.23	.15	4.0R	6.27	.15	3.2R	7.39	.30	1.3X	
21	7.13	.31	1.3	7.12	.18	1.0*	7.26	.27	1.2	A
22	7.07	.19	1.1	7.08	.19	1.1*	7.24	.28	1.4	
23	7.13	.18	1.1	7.16	.18	1.1*	7.28	.28	1.4	PANELS
31	6.63	.18	1.8	6.72	.16	1.4*	7.12	.41	1.9	
32	6.67	.17	1.5	6.69	.17	1.4*	7.02	.43	2.0	
33	6.75	.16	1.5	6.78	.16	1.4*	7.07	.42	1.9	
11	6.37	.29	3.3R	6.38	.28	3.2R				
12	6.29	.29	3.3R	6.29	.29	3.2R				
13	6.40	.28	3.1R	6.41	.27	3.1R				
21	7.13	.31	1.3	7.15	.31	1.3		B		
22	7.07	.32	1.2	7.09	.32	1.2				
23	7.16	.30	1.2	7.15	.31	1.2		PANELS		
31	6.73	.31	2.6	6.74	.30	2.5				
32	6.68	.32	2.8	6.69	.32	2.7				
33	6.78	.30	2.7	6.79	.30	2.8				

R--rejected due to large chi-squared values
X--rejected due to low "validity" (i.e., $V(\Delta)<V(P)+V(C)$) and large constant, K
*--points used in initial average--> $\tau=6.96\pm0.2$ ns

A PANELS	2	5	8	17	19	22	31	(39)
	6	11	18	20	24	27	39	(47)
B PANELS	1	6	11	16	26			
	5	10	15	21	30			

fitting (MHF) and linear interpolation (L-I) were used in order to compare the results of each when used on actual data. Table 4.2 shows that in this case there is very

little difference between the two methods of fitting the cascade, the linear interpolation fits providing only slightly longer lifetime values and slightly lower reduced chi-squared values than the multi-exponential HOMER fits.

The two dimensional analysis(2-D) was the principal method used, but for comparison part of the analysis was done using the multi-dimensional analysis(M-D). The (M-D) analysis produces consistently larger values for the lifetimes (Fig. 4.2) than the (2-D) analysis, although the two agree to within the error estimates.

All the permutations of cascades with primary #1 were rejected due to a large reduced chi-squared. This is the result of the poor quality data of P#1, as attested to by the longer accumulation times. All the other permutations were acceptable according to the rejection criteria described in the previous chapter. Figs. 4.4 and 4.5 are a typical example of the results produced by the computer analysis[1] for this case.

The overall average lifetime was first calculated using only the starred(*) values in Table 4.2, i.e., using only the best fit for each permutation. This gave a value for the lifetime of 6.96±0.2 ns. The results of all the valid analyses were then included in the average and produced essentially the same value 6.99±0.2 ns.

This latter value of the lifetime was then used, along

---

[1] See Appendix II for a description of the computer output.

with a cascade lifetime of 4.49 ns obtained from a HOMER single exponential fit, as initial estimates for a TROY fit, holding the cascade lifetime fixed. Good fits were obtained for primaries #2 and #3 with reduced chi-squared values less than 0.88. This gave a value for the primary lifetime of 7.02±0.22 ns, in excellent agreement with the ANDC result.

Fig. 4.4 Sample analysis(2-D) for F II: variables.

LATENT ROOTS ARE 324.352 -.000 REPEAT=3 DIFF=.0000003

X(I)	Y(I)	VX(I)	VY(I)	VXY(I)	W(I)	D(I)	XX(I)	YY(I)
5.781	10.21	0.123	0.380	0.212	6.07	0.622	5.263	9.267
3.651	8.156	.0182	.0872	.0376	22.4	-.299	3.826	8.573
2.880	8.134	.0061	.0442	.0148	39.3	0.018	2.872	8.111
1.935	7.667	.0146	0.197	.0470	7.97	.0102	1.932	7.654
1.534	7.521	.0043	.0690	.0123	21.2	0.053	1.524	7.457
1.200	7.194	.0041	.0892	.0129	15.9	-.095	1.215	7.307
.8169	7.449	.0019	.0554	.0045	23.9	0.301	0.794	7.103
.4094	6.592	.0022	.0796	.2726	15.9	-.292	0.416	6.921

CENTROID IS 2.1168 7.7442

MATRIX IS 262.7 127.3
127.3 61.7

SLOPE IS 0.485 INTERCEPT IS 6.719

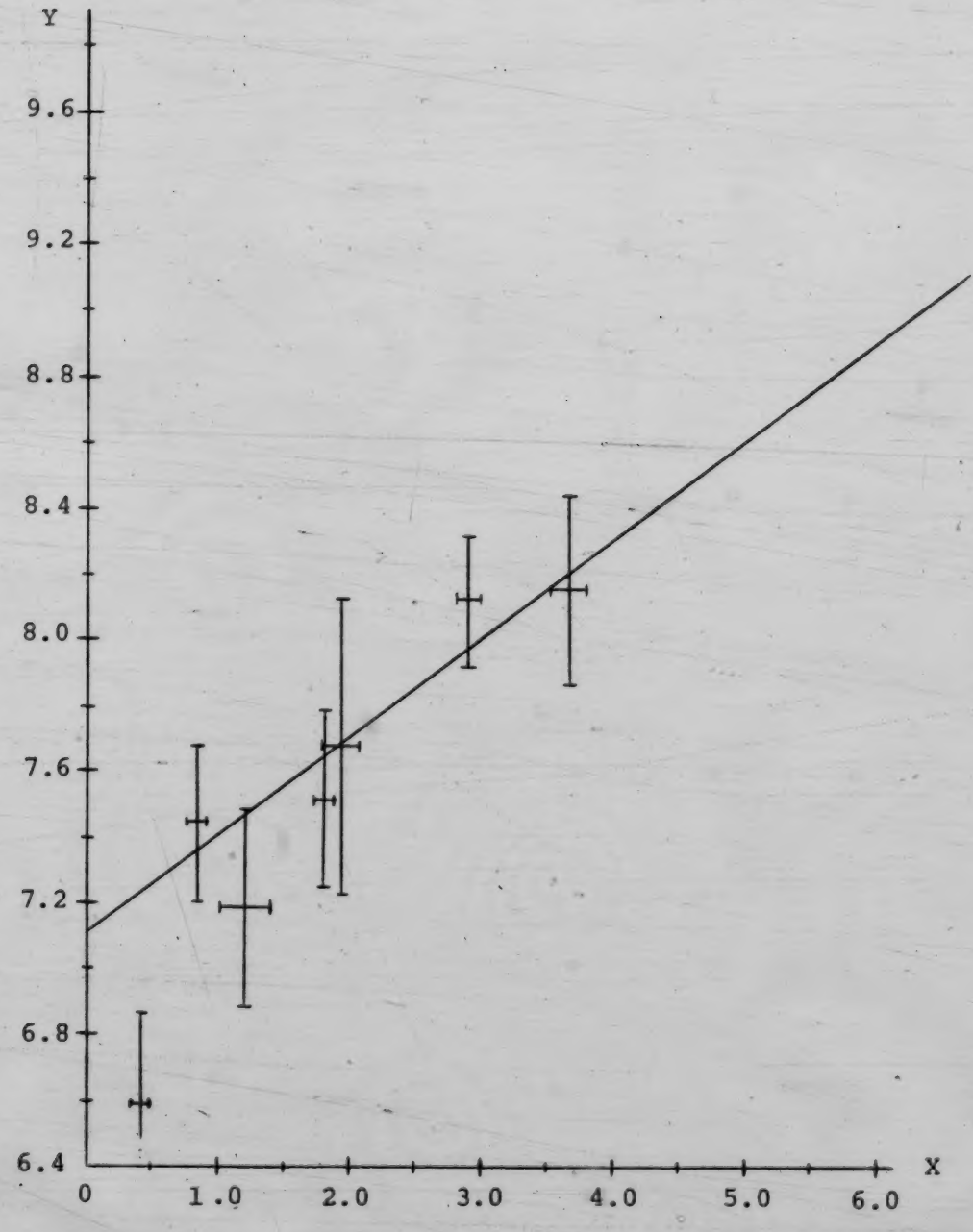
NUMBER OF CYCLES 4

S.D. OF SLOPE=0.064 S.D. OF INT.=0.165

S.D.S. OF CENTROID ARE 0.056 0.104

SQUARED RESIDUALS=8.1 DEGREES OF FREEDOM=6

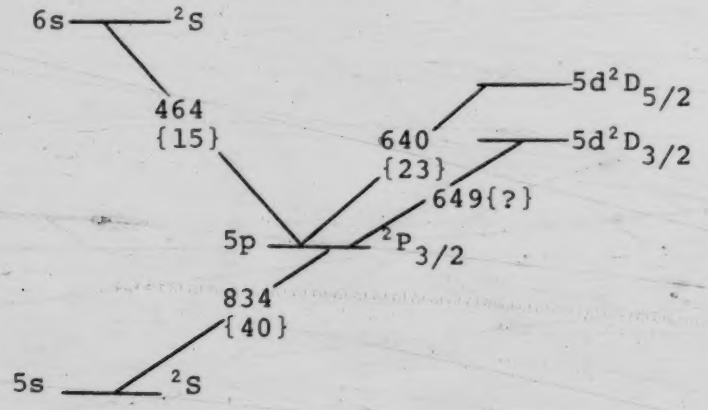
Fig. 4.5 Sample analysis(2-D) for F II: graph.



4.3 Iodine VII: $5p^2P$ Level

This is an example of the use of two direct cascades in ANDC analysis. The relevant part of the decay scheme is shown in Fig. 4.6, and Table 4.3 lists some of the characteristics of the data obtained.

Fig. 4.6 Decay scheme for I VII - $^2P_{3/2}$ level (numbers in {} are approximate intensities).



HOMER fits had to be used for the cascade since all the

Table 4.3 Characteristics of I VII data.

	(A)	#RUNS	PEAK COUNTS	ACCUMULATION TIME(PER POINT)	INTEGRATION LENGTH(NS)
PRIMARY	834	2	3700	18 SEC	0.53
CASCADE	640	2	2000	14 SEC	0.73
CASCADE	464	1	2050	35 SEC	0.66

Beam energy 1.3 MeV Beam velocity=1.537 mm/ns ±1%
Last data point used in analysis occurs at t=4.4 ns.

curves were measured at different distance coordinates. The $^2D_{3/2}$-$^2P_{3/2}$ transition at 649A (Fig. 4.6) was not included in the analysis since the line was severly blended with another stronger line and is also expected to be weak relative to the $^2D_{5/2}$-$^2P_{3/2}$ cascade. Its decay curve should also have a very similar shape to that of the $^2D_{5/2}$-$^2P_{3/2}$ transition.

The transition at 464A (5p-6s) was included in the analysis even though an examination of the spectrum indicates that it probably has a significant blend with the 4f-5g transition. An attempt was made to do the analysis without this line, but all the results had unacceptably large reduced chi-squared values, even though the the lifetimes obtained were close to the those obtained when the 464A line was included in the analysis (this is not unreasonable since this cascade had half the intensity of the 5p-5d cascade). The lifetime of the blend may be such that the shape of this cascade is not significantly changed. The theoretical value of the blend lifetime is 0.04 ns, which is so short compared with the time of the first data point of the cascade (.33ns), that the effect of the blend is probabably negligible.

The lifetimes obtained from the ANDC analysis are shown in Table 4.4. The overall average of these values gives a lifetime of 0.32±.03 ns which is considerably lower than the values obtained from HOMER (0.416±.028) and TROY (.43±.07).

Table 4.4 Results of ANDC analysis on I VII.

PCC #	PANELING A τ	σ	χ^2	PANELING B τ	σ	χ^2	PANELING C τ	σ	χ^2
111	.303	.011	2.7	.340	.020	0.5	.310	.010	1.3*
121	.292	.015	4.7R	.319	.021	3.5	.318	.015	2.1*
211	.337	.013	1.7	.337	.022	2.2	.343	.014	1.8*
221	.325	.017	2.6	.336	.024	4.3R	.356	.017	2.8X

A	1	6	11	16	21	26	31										
PANELS	5	10	15	20	25	30	35										
B	1	5	9	13	17	25	29	33									
PANELS	4	8	12	16	20	25	32	35									
C	3	5	7	9	11	13	15	17	19	21	23	25	27	29	31	33	
PANELS	6	8	10	12	14	16	18	20	22	24	26	28	30	32	34	35	

R--rejected due to large constant and/or chi-squared
X--rejected due to large constant and negative transition probability
*--initial average using best fit of each permutation

* average is 0.32±0.02 (6.3%)

An attempt was made to determine whether the blend in the 464A cascade was having a significant effect on the lifetime by removing a single exponential term from the HOMER fit. The effect of removing the shortest lifetime term (0.24ns) was to give essentially the same lifetime values but with higher reduced chi-squared values for some of the permutations (Table 4.5). The lifetime obtained from the weighted average of the values in Table 4.5 is 0.34±0.01 ns. This value is little different from the previous ANDC value since the entire primary decay curve occurs at t>0.26 ns, i.e., this component of the cascade has a small effect over

Table 4.5 "Blend corrected" ANDC analysis of I VII.

PCC	PANELING A			PANELING C		
	τ	σ	χ^2	τ	σ	χ^2
111	.320	.012	3.0	.331	.010	1.7
121	.294	.016	10.	.329	.016	5.5
211	.353	.014	1.9	.352	.010	1.7
221	.326	.017	6.2	.377	.040	16.

average lifetime = 0.34±0.01 ns

most of the primary decay curve. Fig. 4.7 shows a typical example of the ANDC analysis for this data.

Fig. 4.7 Sample analysis(M-D) for I VII: variables.

I 77 1.3 834 #1
I77 640 #1
I77 464 #1

VAL	FITTED	DELTA	S.DEV	AREA	V(A)	AREA	V(A)	AREA	V(A)
2.9	1356	1349	40	813	46	526	29	445	25
1.1	897	937	29	419	24	124	7	200	11
1.6	444	399	21	218	12	61	4	119	7
1.8	228	228	15	141	10	51	4	105	7
1.9	116	130	12	113	13	51	6	116	13
8.3	39	41	10	65	8	32	4	82	9
3.6	42	38	8	101	29	47	13	136	39

LIFETIME=0.303±0.011

PROBE?	ERRORS
0.9431	0.2608
1.8998	0.2620

CONSTANT K=9.5853±22.3 CHISQR=2.73

Removal of either the middle or largest lifetime component of the cascade resulted in negative transition probabilities and large constants for the ANDC analyses.

The theoretical lifetimes of the two direct cascade levels are about 0.07 ns which, when compared with the relatively long lived cascade decays (the decay curves were measured out to 4.4 ns), indicate that significant cascade effects should be seen in the primary decay. Thus one would expect a significant difference between the ANDC analysis and the multi-exponential fit.

Combination of both the "corrected" and "uncorrected" ANDC results yields a lifetime of 0.33±0.02 ns. The theoretical value[1] of the lifetime, 0.285 +≤20%, agrees with the ANDC result.

4.4 Selenium VI: $4p^2P$ Levels

The Se VI ion has the same number of valence electrons as I VII which was examined in the previous section. Thus the decay scheme shown in Fig. 4.8 is the same as that of Fig. 4.6, except that the principal quantum numbers are lowered by one. However, in this case the cascade transition from the ^2S levels is much weaker than that from the ^2D levels. Furthermore, the data obtained for the $5s^2S$-$4p^2P$ transitions indicate a very short lifetime (<.15ns). Thus only the one direct cascade from the ^2D levels need be included in the ANDC analysis.

These data were taken with ANDC analysis in mind, so

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[1] Obtained by Kwok-tsang Cheng (University of Chicago) using Dirac-Hartree-Fock calculations.

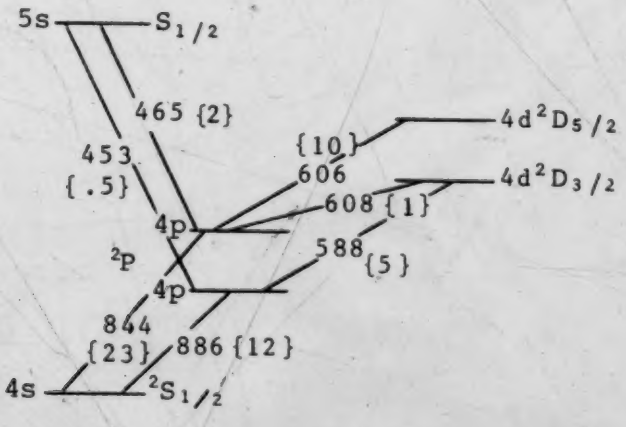
Table 4.6 Characteristics of Se VI Data.

	(A)	NO.	PEAK COUNTS	ACCUMULATION TIME(SEC)
Primary	886	2	3000	60
Cascade	588	2	5000	20
Primary	844	2	3000	30(#1)/60(#2)
Cascade	606	2	4000	25

-last data point taken corresponds to t~11ns
-50 points in each decay curve
-data taken using multi-channel analysis technique

Beam energy 1 MeV Beam velocity 1.55≃mm/ns

Fig. 4.8 Selenium VI decay scheme - $4p\,^2P$ levels
(numbers in {} are approximate measured intensities)



the distance coordinates of the primaries and cascades are matched. However, problems with the foil stepping mechanism affected the distance measurements in some cases, so HOMER fits had to be used for the 886#1 and 844#5 cascades to compensate for this error. Table 4.7 shows the results of

the ANDC analyses using the (2-D), (2-A) and (M-D) methods and using three different partitionings. Note that

Table 4.7 Results of ANDC analysis on Se VI.

PC ##	PANELING A			PANELING B			PANELING C			
844A	τ	σ	χ^2	τ	σ	χ^2	τ	σ	χ^2	
61	.396	.018	0.5	.385	.014	0.5	.379	.014	0.8	|
62	.371	.026	1.1	.377	.014	1.1	.351	.015	1.0	| (2-D)
51	.386	.021	0.7	.371	.017	0.9	.373	.016	0.6	|
52	.345	.019	0.9	.342	.016	1.0	.345	.015	1.5	|
61	.410	.012	1.2	.378	.008	3.7	.391	.010	1.8	|
62	.393	.012	3.4	.376	.008	3.6	.360	.010	2.2	| (M-D)
51		R			R		.351	.012	1.6	|
52	.337	.013	2.5	.325	.007	2.5	.344	.011	0.9	|
51	.340	.017	2.1	.350	.014	1.8	.360	.014	0.6	| (2-A)
52	.336	.017	1.1	.339	.015	1.1	.346	.015	0.5	|
886A										
11	.358	.030	2.1	.364	.028	2.3	.349	.021	1.6	|
12		R			R			R		| (2-D)
31	.424	.017	1.0	.414	.014	1.1	.427	.016	0.8	|
32	.409	.018	1.1	.404	.014	1.4	.414	.016	1.6	|
31	.411	.011	4.2R	.400	.009	10.R	.436	.012	1.6	| (M-D)
32	.382	.010	1.6	.390	.009	4.4R	.401	.012	2.0	|
11		R		.362	.026	2.6	.351	.021	1.8	| (2-A)
12		R			R			R		|

A PANELS	1	6	11	16	21	26	31	36	41	46								
	5	10	15	20	25	30	35	40	45	50								
B PANELS	1	5	10	15	20	25	30	36	43									
	5	10	15	20	25	30	38	46	50									
C PANELS	2	4	6	8	10	12	14	16	18	20	22	24	26	28	30	32	34	
	5	7	9	11	13	15	17	19	21	23	25	27	29	31	33	35	37	

partitioning C is considerably different than A or B in that there is a large degree of overlap between the adjacent panels and 13 points have been dropped from the tail of the decay curve. This was done due to the poor statistics of the last 13 points. The lifetimes obtained from partitioning C are not significantly different from those of A or B, but there is a noticeable improvement in the reduced chi-squared of some of the permutations. This indicates that the choice of paneling does not affect the lifetime, but may affect the statistics of the analysis. Figs. 4.9 and 4.10 show a typical example for this analysis.

The difficulty in analyzing the permutations with 886#1 may be due to inadequate multi-exponential fits or simply poor data sets. A correction of the beam velocity to 1.576 mm/ns ±2% lowers the lifetimes so that the average values are 0.357±0.029 (844A) and 0.401±0.029 ns (886A). The ratio of the two lifetimes is 0.89±11%. Theoretically this ratio should equal the cube of the ratio of the corresponding wavelengths, which is 0.86. The results of TROY multi-exponential analysis yields the same lifetime for both levels: 0.52±0.01 ns.

4.5 Summary

This chapter demonstrated the practical application of ANDC analysis to actual experimental data. An important consideration when performing an experiment is the integration length. If ANDC is to be applied to the data,

Fig. 4.9 Sample analysis (2-A) for Se VI: graph.

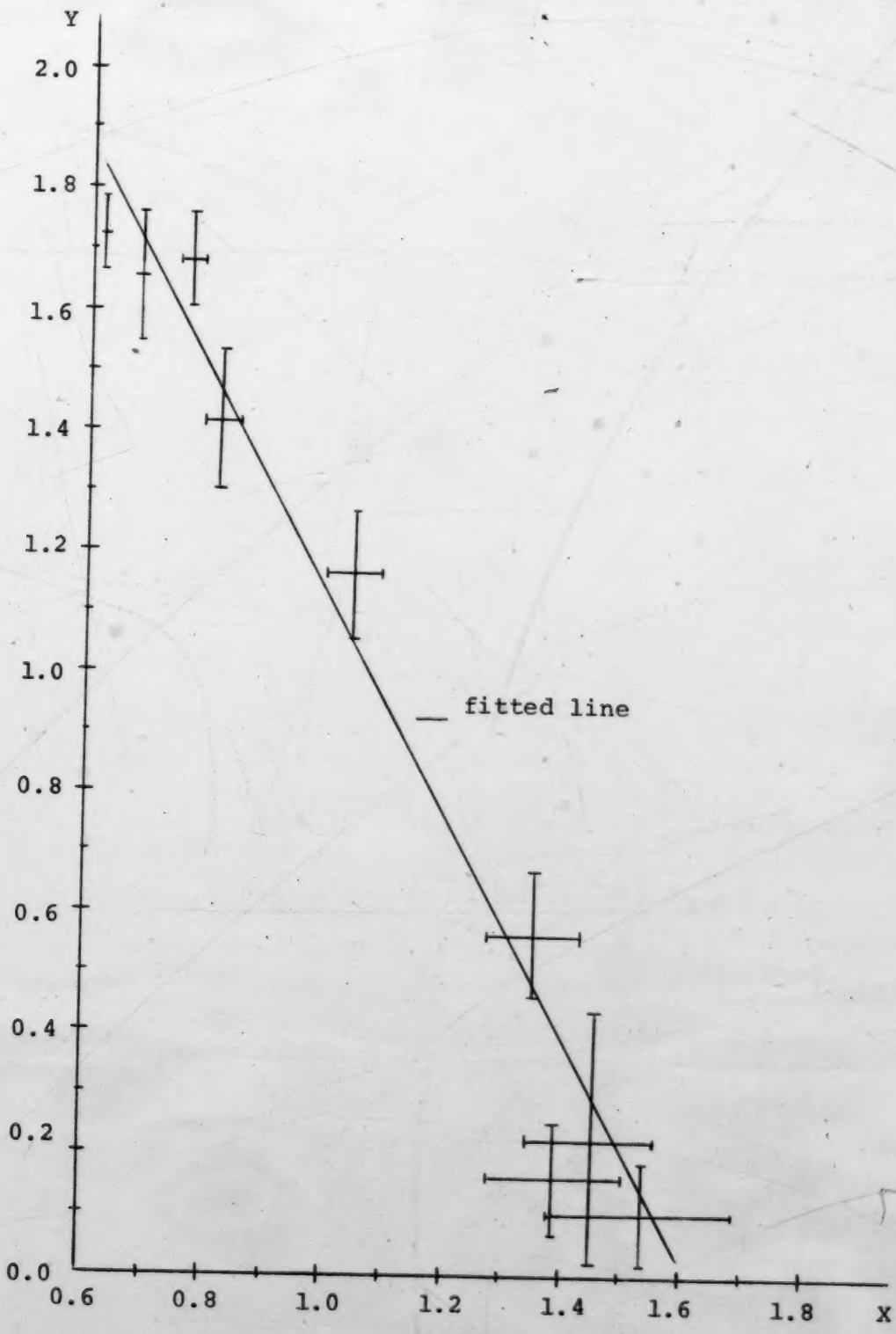


Fig. 4.10 Sample analysis(2-D) for Se VI: variables.

LATENT ROOTS ARE -0.000 647.7 REPEAT=4 DIFF=.0000002

X(I)	Y(I)	VX(I)	VY(I)	VXY(I)	W(I)	D(I)	XX(I)	YY(I)
0.761	1.682	.0001	.0034	.00012	1033	.0511	0.753	1.591
0.616	1.723	.0002	.0061	.00017	607	-.057	0.624	1.829
0.677	1.654	.0003	.0108	.00031	337	-.037	0.683	1.721
0.819	1.415	.0007	.0125	.00056	259	-.026	0.825	1.459
1.044	1.164	.0021	.0110	.00118	197	.0522	1.020	1.099
1.346	0.568	.0060	.0107	.00146	121	.0331	1.322	0.542
1.450	0.229	.0102	.0579	.00095	46	-.037	1.466	0.277
1.387	0.162	.0122	.0083	.00083	84	-.126	1.502	0.211
1.534	0.102	.0226	.0072	.00091	50	-.024	1.558	0.107

CENTROID IS 0.815 1.476

MATRIX IS 147.4 -271.5
-271.5 500.4

SLOPE IS -1.843 INTERCEPT IS 2.979

NUMBER OF CYCLES=3

S.D. OF SLOPE=0.174 S.D. OF INTERCEPT=0.148

S.D.S. OF CENTROID 0.0111 0.0363

SQUARED RESIDUALS=7.4 DEGREES OF FREEDOM=7

all decay curves that are to be used in a given analysis should either have the same integration length or the integration length should be so small as to have a negligible effect on all the decay curves.

The lifetime of the $3p\,^3D$ level in F II was determined to be 6.99±0.34 ns (including the uncertainty in the velocity) by using ANDC analysis. As this analysis gave no reason to suspect any problems such as undetected blends or missing direct cascades and the result is also in excellent agreement with the conventional multi-exponential analysis,

one can be confident in the lifetime obtained.

In the case of I VII, the lifetime of the $5p^2P_{3/2}$ level was determined to be 0.33±0.33 ns by ANDC analysis. However, even though the ANDC analysis is self-consistent, there is some doubt of the validity of this result due to the blend present in the direct cascade at 464A. This result is also significantly lower than that given by multi-exponential analysis, although it agrees with the theoretical value. Analysis of further data, with the blend at least partially resolved, is necessary to obtain a conclusive value for the lifetime of this level.

The lifetimes of the $4p^2P_{3/2}$ and $4p^2P_{1/2}$ levels of Se VI were determined by ANDC analysis to be 0.357±0.029 ns and 0.401±0.029 ns respectively. The analyses were such that confidence can be placed in this first value. The latter value is not as certain due to the problems of in one of the two primary runs analyzed (one should be wary of drawing conclusions from only one "good" primary). However, since the lifetimes have the correct theoretical ratio, the second lifetime is probably correct also. As in the previous case the ANDC lifetimes are much less than those obtained by conventional analysis.

CHAPTER V

CONCLUSION

The particular method of applying ANDC analysis developed in this work has shown itself to be a satisfactory means to determine lifetimes in decay situations involving significant cascading. It has particular value where multi-exponential techniques fail to give satisfactory results, but is also useful as a check on the results of multi-exponential analysis.

In the highly cascaded synthetic data situations of Chapter III, the multi-exponential method proved to be inadequate, giving either misleading results (ie. actual lifetimes not within one standard deviation of the determined value) or results which were only correct by virtue of very large error estimates (±20%). The ANDC lifetimes on the other hand were accurate to well within reasonable error limits (<7% in all cases). One of the synthetic data cases also showed that cascade effects will not necessarily cause the lifetime obtained by multi-exponential analysis to be too large, as had been previously accepted in the past.

The analyses of the real data of Chapter IV indicated that there is a point of diminishing returns in including data points from the tail of a decay curve where there are fewer than about 100 counts per data point, since the statistical fluctuations will then exceed 10% of the signal, causing the error in the intensity differences, Δ, to be

very large. The weighting procedures in the fitting routine insure that these points will have a small effect on the lifetime, but their presence certainly does not improve the analysis.

Both the synthetic and real data cases indicate the desirablity of using as many different primary runs as possible in the ANDC analysis. Since the primary data set dominates the determination of the lifetimes and error estimates, this practice produces a number of independent lifetime measurements which can then be averaged. Three primary data sets would appear to be the minimum number to be collected for a given transition. In the case where one data set proves to be unsatisfactory in the analysis, this would still leave two data sets to provide two independent ANDC analyses whose results can be checked for consistency, and then averaged. Similarly, it would also be wise to obtain at least two data sets for each direct cascade.

Before collecting the decay curve data, the decay scheme and the spectrum (at the energy at which the decay data is to be taken) should be studied carefully to determine all the possible significant direct cascades which should be included in the ANDC analysis. At the same time, the possibility of blends in any of these decays should be considered. It is also necessary that all the decay curves to be used in a given ANDC analysis be taken at the same beam energy to insure that the level populations are the same for each decay curve (a basic assumption in ANDC

analysis). As indicated in Chapter IV, it is also necessary that the optical integration length along the beam be the same for all decay curves or that it be negligible for all decay curves in a particular ANDC analysis.

The computer program developed allows the ANDC analysis to be performed several different ways (see Table 5.1).

Table 5.1 Variations possible in the computer program used for ANDC analysis.

a) FITTING PROCEDURES

EQUATION	VARIABLES	NAME OF ANALYSIS
$P/\Delta = \tau + \xi C/\Delta$	$Y = P/\Delta, X = C/\Delta$	(2-D)
$\Delta/P = 1/\tau - \xi C/P$	$Y = \Delta/P, X = C/P$	(2-A)
$\Delta = P/\tau - \sum_i \xi_i C/\tau$	$Z = \Delta, Y = P, X_i = C_i$	(M-D)

b) FORM OF DECAY CURVE DATA

CURVE	FORM	DESCRIPTION
PRIMARY	-REQUIRES ORIGINAL HOMER FORMAT DATA SETS	NORMAL MODE
CASCADE	-ORIGINAL HOMER FORMAT WITH MATCHING DISTANCE VARIABLES	NORMAL MODE
CASCADE	-ORIGINAL HOMER FORMAT WITH MIS-MATCHED DISTANCE VARIABLES	LINEAR INTERPOLATION (L-I) MODE
CASCADE	-SIX PARAMETER EXPONENTIAL FIT TO DECAY CURVE	MULTI-EXPONENTIAL HOMER FIT (MHF) MODE

*** note that all the cascades for a given ANDC analysis must be input in the same mode.

c) Analysis can be done using any desired panel division of the decay curves.

Previous chapters have discussed the relative merits of each variation on the method. However, since all the variations are capable of producing a "valid" ANDC analysis, performing the same analysis using a few different variations is a useful check on the self-consistency of the results. Variations of the paneling does not usually affect the lifetime significantly but can give a better fit in the sense of lower error estimates and/or reduced chi-squared values.

The results of an ANDC analysis may be accepted as valid subject to the satisfaction of the rejection criteria on various quantities resulting from the analysis which were discussed in sec. 3.4. Once an acceptable lifetime has been obtained for a particular level (following all the procedures outlined above), it is of interest to attempt a multi-exponetial fit using the determined primary lifetime and the cascade lifetimes (as determined by HOMER fits) as initial estimates for the fit (cf. sec. 3.3), possibly constraining one or more of the fitting parameters. Frequently, the lifetime determined by the multi-exponential fit will agree with the ANDC lifetime, indicating that the decay situation was not so severely cascaded as to render conventional procedures inadequate. This will also serve as a check on the error estimate of the lifetime. In the cases where the multi-exponential fit and ANDC lifetimes are inconsistent, the ANDC lifetime is to be preferred. The only reason an acceptable ANDC analysis would be incorrect

is when a significant direct cascade had been omitted or an undetected blend had occurred in either the primary or one of the direct cascades. As was discussed in sec. 3.4, there is no absolute guarantee that the application of the rejection criteria will always detect these possible problems. However, these situations are less likely than the possibility of some difficulty with the multi-exponential analysis (cf. sec. 3.3).

Refinements in the application of the ANDC method can be made by further study of synthetic data situations. The relationship between blends and the determined values for the lifetimes, estimated errors and the rejection criteria can be studied by varying the type and amount of blend present. A less qualitative "curvature" rejection criterion might be developed by calculating the directional deviation of the fitted curve from the actual ANDC points. Also, such studies could place definite limits on what constitutes a "significant" direct cascade. The relationship between the number of counts (peak and minimum) and the quality of the resulting ANDC analysis could be easily studied using synthetic data situations. This would be of value in determining how many counts one needs and how far into the decay curve tail on must go in order to obtain an optimal data set for ANDC analysis.

In the original paper describing the ANDC method (CBB 70), it was stated that the method was "very promising". However, since then, very little use has been

made of this technique due to an apparent lack of success in its practical application. The results of this work show that the ANDC method is able to fulfill its original promise.

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APPENDIX I

Modeling of Cascaded Decay Schemes

The population of a particular level in a known decay scheme can be determined as a function of time if all the relevant initial populations and transition probabilities are known. This is done by solving the coupled differential equations governing the decays of each state, as was done for the three level system in sec. 1.3. The equations have been solved and put in mnemonic form by L.J. Curtis (Cu 68) for a generalized decay scheme. The formula which follows refers to a decay scheme in which the energy levels are labelled from 1 to m in order of increasing energy. The population of level n is then given by

$$N_n(t) = N_n(0)e^{-\alpha_n t} + \sum_{i=n+1}^{m} \{i\to n\} + \sum_{i=n+1}^{m-1} \sum_{j=i+1}^{m} \{j\to i\to n\} + \sum_{i=n+1}^{m-2} \sum_{j=i+1}^{m-1} \sum_{k=j+1}^{m} \{k\to j\to i\to n\} + \ldots + \sum\{m\to \ldots \to n\}, \tag{A1.1}$$

where the brackets correspond to contributions from single stage, two stage, three stage etc. transitions into the state n, and

$$\{i\to n\} = N_i(0)A_{in}\left[\frac{e^{-\alpha_i t}}{(\alpha_n-\alpha_i)} + \frac{e^{-\alpha_n t}}{(\alpha_i-\alpha_n)}\right]$$

$$\{j\to i\to n\} = N_j(0)A_{ji}A_{in}\left[\frac{e^{-\alpha_j t}}{(\alpha_i-\alpha_j)(\alpha_n-\alpha_j)} + \begin{array}{c}\text{cyclic permutations}\\ \text{of } \{j,i,n\}\end{array}\right]$$

$$\{k\to j\to i\to n\} = N_k(0)A_{kj}A_{ji}A_{in}\left[\frac{e^{-\alpha_k t}}{(\alpha_j-\alpha_k)(\alpha_i-\alpha_k)(\alpha_n-\alpha_k)} + \text{cyclic perms}\right]$$

$$\{m\to\ldots\to n\} = N_m(0)\left\{\prod_{i=n}^{m-1} A_{i+1,i}\right\}\left[\sum_{j=n}^{m}\frac{e^{-\alpha t}}{\prod_{\substack{k=n\\k\neq j}}^{m}(\alpha_k-\alpha_j)}\right].$$

When modeling cascade decay schemes for the purpose of lifetime determination, it is more convenient to have the level populations expressed in the form

$$N_n(t) = \sum_i D_i e^{-\alpha_i t} .$$

This can easily be achieved by factoring eqn. A1.1 into

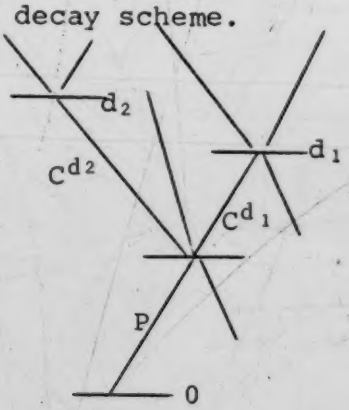
$$N_n(t) = \sum_{r=n}^{m}\left\{\sum_{j=r}^{m} N_j(0)\frac{\prod_{i=n}^{j-1} A_{i+1,i}}{\prod_{\substack{k=n\\k\neq r}}^{m}(\alpha_k-\alpha_r)}\right\}e^{-\alpha_r t} = \sum_{r=d}^{m} Q_{rn}\, e^{-\alpha_r t} .$$

Thus if one has a decay scheme as shown in Fig. A1.1, one can then write for the intensities of the primary and direct cascades,

$P(t)= \sum_{r=n}^{m} P_r e^{-\alpha_r t}$ and $C^d(t) = \sum_{r=d}^{m} C_r^d e^{-\alpha_r t}$, where

$P_r = A_{n0}Q_{rn}$ and $C_r^d = A_{dn}Q_{rd}$. It should be noted that the index, d, refers to a particular cascade path, so that the same level may occur in more than one path.

Fig. A1.1 Generalized decay scheme.



APPENDIX II

A description of the input parameters and their format are contained at the beginning of the listing for the main program. The two borrowed subroutines, REGRES and CORREG, and the simple plotting routine, APLOT, are not listed here.

The program output consists of a partial listing of the input parameters (not including data sets) and the results of the analysis, samples of which were given in Chapter IV.

The results of (2-D) or (2-A) analyses[1], consist of

- the calculated ANDC variables(X(I) and Y(I)) with a plot of these variables (if desired),
- the calculated variances and covariance (VX,VY,VXY) of the variables,
- the weight(W) calculated for each point,
- a measure of the deviation(D) of the fitted line from the calculated variables,
- XX(I),YY(I), the fitted values for the variables,
- the center of "mass"(CENTROID) of the calculated variables,
- the parameters of the fit (SLOPE and INTERCEPT), where the intercept is the lifetime(2-D) or its inverse(2-A),
- the standard deviation(S.D.) of the CENTROID, SLOPE and INTERCEPT,
- the SQUARED RESIDUALS and # DEGREES OF FREEDOM, the ratio of which is the reduced chi-squared,
- other output related to the details of the computation (MATRIX, LATENT ROOTS etc.).

---

[1] For a description of the mathematical details of this fitting procedure see CRRW 72.

The output of the (M-D) analysis consists of a check on the validity of the assumption that has a significantly larger error than any of the areas,

$VAL=(V(\Delta)/\Delta^2)/\ (V(A_i)/A_i^2$, and

-the ANDC variables DELTA and AREAs ,

-the FITTED value for DELTA,

-the LIFETIME and its estimated standard deviation,

-the transition probability(PROBE?) modified by the efficiency for each direct cascade,

-the value of the constant, K, and its estimated error,

-the value of the reduced CHI-SQUARED.

The following pages contain a listing of the main program used for the ANDC analyis and the subroutine which does linear interpolation for mis-matched (distances) cascade data sets.

```
C   PROGRAM FOR ANDC ANALYSIS OF DECAY CURVES
C
C   DEVICE 7    HOMER PARAMETERS TO RECONSTRUCT CASCADE(S)
C       LINE N AMP,TL,ALIGN,ICOM     (7F9.4,4A4)
C   AMP=AMPLITUDES TL=LIFETIMES ALIGN= INITIAL DISTANCE OF
C                                      CASCADE FIT
C   DEVICE 8    DECAY DATA IN HOMER FORMAT (P'S THEN C'S)
C   DEVICE 9    CONTROL DATA
C      LINE 1   IND,ND,IFLAG,IM,IS,IF,NC,NPERM,VEL
C                (4I1,2I3,1X,4I1,I3,F6.1)
C       LINE 2  PI              (20I3)
C       LINE 3  PF              (20I3)
C       LINE 4/5/6 ID(1,I)                    (30I2)
C       LINE 5/6/7 ID(2,I)
C       LINE 6/7/8 ID(3,I)     IF APPLICABLE
C       LINE 7/8/9 ID(4,I)      IF APPLICABLE
C   IND----HOMER DEPENDENT/INDEPENDENT/INTERPOLATION-1/2/3
C   ND-----DISPLAY PARAMETERS
C          1--NO PLOT,SHORT STATISTICS
C          2--PLOT,SHORT STATS.
C          3--NO PLOT,LONG STATS.
C          4--PLOT,LONG STATS.
C   IFLAG--(2-D)/(M-D)/(2-A) --0/1/2
C   IM-----NUMBER OF DECAY CURVES IN ONE ANALYSIS
C   IS-----FIRST DATA POINT OF PRIMARY USED IN ANALYSIS
C   IF-----LAST DATA POINT TO BE USED IN ANALYSIS
C          WHEN IF=0: PI,PF ARE SPECIFIED BY USER
C   NC-----NO. OF DECAYS OF EACH TYPE (IE.#P,#C1,#C2,#C3)
C   NPERM--NO. OF PERMUTATIONS DESIRED  MAX=40
C          =0 =>AUTO PERMUTATION
C   VEL----BEAM VELOCITY (MM/NS)
C   PI-----INITIAL PANEL POSITIONS(MUST HAVE PI(1) SMALLEST)
C   PF-----FINAL PANEL POSITIONS (LAST MUST BE LARGEST)
C   ICOM---ONE COMMENT FOR EACH DECAY CURVE (MAX=20)
C   ID-----PERMUTATIONS: NOT NECESSARY IF AUTO-PERM ON
C        CAUTION: # OF POSSIBLE PERMS MAY BE > MAX. ALLOWED.
C     1ST LINE   PRIMARY ID'S      1 2 3 4
C     2ND LINE   CASCADE ID'S      2 3 2 3
C              ETC.
C
C   SUBPROGRAMS REQUIRED TO BE IN USERS FILE:
C    REGRES CORREG APLOT INTER
C
C   COMMENTS:  INTERPOLATION SHOULD BE USED WITH ONLY ONE
C   PRIMARY AT A TIME (UNLESS P'S DISTANCES MATCH)
C
      REAL*8 XX(20),YY(20),VX(20),VY(20),VXY(20)
     1,SUM,SUMX,SUMY,SUMXY,SUMX2,SUMY2,Z,ZX,ZY,XI,YI
      REAL Y(80),D(80),TA(80),AMP(3),TL(3),T(80),YFIT(20),
     1SDT(200),YA(400),DEL(200),SYA(400),SD(200),R(40),A(4),
     2SIGA(4)
      INTEGER ICOM(4,20),ILO(3),NPP(20),ID(4,40),IHOM(5,3)
     1,PI(21),PF(20),NPNP(5),NC(4),ICOML(4,4),ICOM1(4)
```

```
      COMMON /IDISP/ND /AREA/YA,NPAN,NPNP
      EQUIVALENCE (XX(1),Y(1)),(YY(1),Y(41)),(VX(1),D(1)),
     1(VY(1),D(41)),(TL(1),TA(44)),(ILO(1),TA(47))
     1,(VXY(1),TA(1)),(AMP(1),TA(41))
     2,(NPP(1),PI(1)),(NC(1),NP),(NC(1),NPNP(2))
     3 ,(NC1,NC(2)),(NC2,NC(3)),(NC3,NC(4))
      DATA IHOM/'  HO','MER ','DEPE','NDEN','T   ',
     1'  HO','MER ','INDE','PEND','ENT ','LINE','AR I'
     2,'NTER','POLA','TION'/
      CALL TRACER(-1)
 1000 READ(9,100,END=2000)IND,ND,IFLAG,IM,IS,IF,NC,NPERM,VEL
      PI(21)=0
      IF(IF.NE.0)GO TO 73
C
C   PANEL SIZES
C
      READ(9,106)PI,PF
      DO 71 J=3,20
      IF(PI(J).EQ.0)GO TO 72
   71 CONTINUE
   72 NPAN=J-1
      GO TO 77
C
C   AUTOMATIC PANELING
C
   73 P=IS+4
      DO 74 J=1,20
      PI(J)=P-4
      PF(J)=P
      P=P+5
      IF(P.GT.IF)GO TO 75
   74 CONTINUE
   75 NPAN=J
      J=J+1
      DO 76 K=J,20
      PI(K)=0
   76 PF(K)=0
   77 WRITE(6,150)(IHOM(K,IND),K=1,5),PI,
      NS=PF(NPAN)
      IDUP=NS-1-PI(1)
      NPAC=NP+NC(2)+NC(3)+NC(4)
C
C   AUTOMATIC PERMUTING OF PRIMARIES AND CASCADES
C
      IF(NPERM.NE.0)GO TO 13
      K=0
      DO 60 I=1,NP
      DO 60 J=1,NC1
      DO 60 L=1,NC2
      DO 60 M=1,NC3
      K=K+1
      ID(1,K)=I
      ID(2,K)=J
```

```
      ID(3,K)=L
      ID(4,K)=M
   60 CONTINUE
      NPERM=K
      IF(NPERM.GT.40)GO TO 2000
      GO TO 78
   13 DO 700 I=1,IM
      READ(9,102)(ID(I,J),J=1,NPERM)
  700 CONTINUE
   78 M=0
      NPNP(1)=0
C
C   READ IN DECAY CURVES
C
      DO 14 I=1,NPAC
      IF(I.LE.NP)GO TO 1500
      GO TO (50,1500,3000),IND
 1500 READ(8,103)(ICOM(J,I),J=1,4),N,BGR
      READ(8,104)(D(J),Y(J),TA(J),J=1,N)
      DO 1 J=1,N
    1 Y(J)=Y(J) -TA(J)*BGR
C
C   MATCH DISTANCE ENDPOINTS
C
      IF(I.GT.1)GO TO 5
      DS=D(IS)
      L=0
      DO 2 K=IS,N
      L=L+1
    2 T(L)=(D(K) -DS)/VEL
      GO TO 6
    5 DO 3 K=1,N
      IF(D(K).EQ.DS)GO TO 4
    3 CONTINUE
    4 IS=K
    6 IF(IS.EQ.1)GO TO 10
      L=0
      DO 7 K=IS,N
      L=L+1
    7 Y(L)=Y(K)
      GO TO 10
C
C   CALCULATE CASCADE AMPLITUDES FROM HOM  FIT
C
   50 READ(7,105)AMP,TL,ALIGN,(ICOM(J,I),J=1,4)
      ALIGN=(ALIGN-DS)/VEL
      DO 56 K=1,3
      TLT=TL(K)
      ILO(K)=1
      IF(TL(K).EQ.0.0)GO TO 57
      AMP(K)=AMP(K)*EXP(ALIGN/TLT)
      GO TO 56
   57 ILO(K)=3
```

```
   56 CONTINUE
      DO 55 K=1,NS
      B=0.0
      DO 54 J=1,3
      IBB=ILO(J)
      GO TO (51,54,53),IBB
   51 TLT=TL(J)
      E=-T(K)/TLT
      IF(E.GT.-35.)GO TO 52
      ILO(J)=2
      GO TO 54
   52 B=AMP(J)*EXP(E)+B
      GO TO 54
   53 B=B+AMP(J)
   54 CONTINUE
   55 Y(K)=B
      WRITE(6,152) (ICOM(K,I),K=1,4),AMP,  ,ALIGN
      GO TO 10
 3000 CALL INTER(N,DS,D,Y,ICOM1)
      DO 3001 J=1,4
 3001 ICOM(J,I)=ICOM1(J)
C
C   CALCULATE AREAS AND DELTAS
C
   10 NL=N-NS
      WRITE(6,151) (ICOM(K,I),K=1,4),IS,  ,NPAN,VEL
      JJ=0
      DO 12 J=1,NPAN
      K1=PI(J) -PI(1)+1
      K2=PF(J) -PI(1)+1
  222 K2M1=K2-1
      AA=0.0
      SA=0.0
      DT1=T(K1+1) -T(K1)
      DO 11 L=K1,K2M1
      DT=T(L+1) -T(L)
      AA=AA+(Y(L)+Y(L+1))*DT
      IF(L.EQ.K1)GO TO 11
      SA=SA+Y(L)*(DT+DT0)**2
   11 DT0=DT
      SA=(SA+DT1*DT1*Y(K1)+Y(K2)*DT*DT)/4.
      M=M+1
      YA(M)=AA/2.0
      IF(I.GT.NP)GO TO 12
      Q=0.
      SDP=0.
      IDT=1
      TD=T(K1)
      KC=K1-3
C   DO LEAST SQUARES FIT FOR DELTA
  211 SUM=0.
      SUMX=0.
      SUMY=0.
```

```
      SUMXY=0.
      SUMX2=0.
      SUMY2=0.
      IGO=0
      IF(K1.LT.4.AND.IDT.EQ.1) IGO=2
      IF(K2.GT.IDUP.AND.IDT.EQ.-1) IGO=-2
  221 DO 250 II=1,5
      L=KC+II+IGO
      XI=T(L)
      Z=Y(L)
      YI=DLOG(Z)
      ZY=Z*YI
      ZX=Z*XI
      SUM=SUM+Z
      SUMX=SUMX+ZX
      SUMY=SUMY+ZY
      SUMX2=SUMX2+ZX*XI
      SUMXY=SUMXY+ZY*XI
      SUMY2=SUMY2+ZY*YI
  250 CONTINUE
      DIV=SUM*SUMX2-SUMX*SUMX
      CA=(SUMX2*SUMY-SUMX*SUMXY)/DIV
      Z=SUMXY*SUM-SUMX*SUMY
      CB=Z/DIV
      JJ=JJ+1
      R(JJ)=DABS(Z/DSQRT(DIV*(SUM*SUMY2-SUMY*SUMY)) )
C     CALCULATE DELTA
  251 P=EXP(TD*CB+CA)
      SDP=SDP+P/(R(JJ)**2)
      Q=Q+IDT*P
      IF(IDT.LT.0) GO TO 260
      TD=T(K2)
      IDT=-IDT
      KC=K2-3
      GO TO 211
  260 CONTINUE
C
      DEL(M)=Q
      SD(M)=SDP/4.
      SDT(M)=SQRT(SD(M))
      R(J)=(R(JJ)+R(JJ-1))/2.
   12 SYA(M)=SA
      IF(I.GT.NP)GO TO 14
      JJ=JJ/2
      WRITE(6,154) (R(L),L=1,JJ)
   14 CONTINUE
C
      IF(IFLAG.NE.1)GO TO 17
C
C     (M-D) ANDC ANALYSIS
C
      DO 18 I=1,NPERM
      WRITE(6,157)
```

```
      L=NPAN*(ID(1,I) -1)+1
      CALL REGRES(A,DEL(L),SDT(L),NPAN,IM,ID(1,I),1,YFIT,
     1A0,A,SIGA0,SIGA,R,RMUL,CHISQR,FTEST)
      IN=0
      DO 23 M=1,IM
      NN=ID(M,I)+IN
      IN=IN+NC(M)
      DO 24 J=1,4
   24 ICOML(J,M)=ICOM(J,NN)
      WRITE(6,160) (ICOM(J,NN),J=1,4)
   23 CONTINUE
  160 FORMAT(' ',4A4)
      WRITE(6,162)
  162 FORMAT('-','VALIDITY',2X,'FITTED',  ,'DELTA ',2X,
     1'S.DEV ', 4(4X,'AREA',4X,'VARIANCE  )
      BOD=0.0
      DO 20 J=1,NPAN
      AOD=0.0
      DO 21 K=1,IM
      M=NUM(J,K,ID(1,I))
      Y(K)=YA(M)
      D(K)=SYA(M)
   21 AOD=AOD+D(K)/(Y(K)**2)
      K=L+J-1
      AOD=(SDT(K)/DEL(K))**2/AOD
      WRITE(6,161) AOD,YFIT(J),DEL(K),SDT(K),(Y(K),D(K),
     1K=1,IM)
  161 FORMAT(1X,F6.1,1X,3(2X,F6.0),8(2X,F8.1))
   20 CONTINUE
      TAU=1./A(1)
      DTAU=SIGA(1)*TAU*TAU
      DO 1600 J=2,IM
 1600 A(J)=-A(J)
      WRITE(6,164) TAU,DTAU,(A(J),SIGA(J),J=2,IM)
  164 FORMAT(///' LIFETIME=',F7.3,1X,F7.3,16X,' PROBE? ',4X,
     1'ERRORS ',3(/40X,F8.4,3X,F8.4))
      WRITE(6,165) A0,SIGA0,CHISQR
  165 FORMAT('0CONSTANT= ',F9.4,3X,F9.4,5X,'CHISQR=',F8.3)
      WRITE(1,183) ((ICOML(J,K),J=1,4),K=1,IM)
      WRITE(1,180) TAU,CHISQR,A0,(A(J),J=2,IM)
      WRITE(1,182) DTAU,SIGA0,(SIGA(J),J=2,IM)
  180 FORMAT(16X,F7.3,5X,F6.3,5X,F9.4,5X,3(F8.4,1X))
  182 FORMAT(16X,F7.3,16X,F9.4,5X,3(F8.4,1X))
  183 FORMAT(' *',20A4)
   18 CONTINUE
      GO TO 1000
C
C   (2-D) ANDC ANALYSIS
C
   17 DO 16 I=1,NPERM
      L=NPAN*(ID(1,I) -1)
      M=NPAN*(ID(2,I)+NP-1)
      DO 15 J=1,NPAN
```

```
      IP=L+J
      IC=M+J
C
C   ALTERNATE (2-D) METHOD
C
      IF(IFLAG.EQ.0)GO TO 8000
      Q=YA(IP)
      D2=Q*Q
      SDP=SYA(IP)
      AOD=DEL(IP)/Q
      VY(J)=(SD(IP)+AOD*AOD*SDP)/D2
      GO TO 8001
C
 8000 SDP=SD(IP)
      Q=DEL(IP)
      D2=Q*Q
      AOD=YA(IP)/Q
      VY(J)=(SYA(IP)+AOD*AOD*SDP)/D2
 8001 BOD=YA(IC)/Q
      VX(J)=(SYA(IC)+BOD*BOD*SDP)/D2
      VXY(J)=AOD*BOD*SDP/D2
      YY(J)=AOD
   15 XX(J)=BOD
      T1=ID(1,I)
      T2=ID(2,I)+NP
      WRITE(6,153)(ICOM(K,T1),K=1,4),(ICOM(K,T2),K=1,4),
     1IS,IF,NPAN
      GO TO (601,602,601,602),ND
  602 CALL APLOT(NPAN,XX,YY)
      WRITE(6,157)
  601 CALL CORREG(NPAN,XX,YY,VX,VY,VXY)
   16 CONTINUE
      GO TO 1000
  100 FORMAT(4I1,2I3,1X,4I1,I3,F6.1)
  102  FORMAT(40I2)
  103 FORMAT(4A4,54X,I2/16X,F6.3)
  104 FORMAT(5(F3.1,F6.0,F4.1,1X))
  105 FORMAT(7F9.3,4A4)
  106 FORMAT(21I3/20I3)
  150 FORMAT('1',10X,5A4//10X,' PTS. IN PANELS ARE '
     1/21I3/20I3//)
  151 FORMAT(10X,4A4,'IS=',I3,2X,'IF=',I3,2X,'NPAN=',I3,2X,
     'VEL=',F5.3)
  152 FORMAT(10X,4A4,'  AMPLITUDES',3F10.2,' LIFETIMES',
     2 3F8.3,3X,'ALIGN=',F5.2)
  153 FORMAT('1',4A4,' AND ',4A4,'  IS=',I3,2X,'IF=',I3,2X,

     2'NPAN=',I3,I3//)
  154 FORMAT(' LIN CORR.COEFF.'/1X,20('*',F5.2))
  157 FORMAT('1')
 2000 STOP
      END
      FUNCTION FCTN(Z,I,J,NX)
```

```
      DIMENSION NPNP(1),YA(400),NX(1)
      COMMON /AREA/YA,NPAN,NPNP
      M=NUM(I,J,NX)
      FCTN=YA(M)
      RETURN
      END
      FUNCTION NUM(I,J,N)
      DIMENSION N(1),YA(400),NPNP(5)
      COMMON /AREA/YA,NPAN,NPNP
      M=0
      K=N(J)
      DO 1 L=1,J
    1 M=M+NPNP(L)
      NUM=NPAN*(K+M-1)+I
      RETURN
      END
      SUBROUTINE INTER(NP,DS,X,YC,ICOM1)
C
C
C     THIS PROGRAM READS THE CASCADE DATA SET AND
C     CALCULATES THE AMPLITUDES
C     AT THE DISTANCE COORDINATES GIVEN BY THE PRIMARY
C     (LINEAR INTERPOLATION)
C
      REAL TA(80),YC(80),X(80)
      INTEGER ICOM1(4),ICOM2(4),IX(80),IXC(80),IYC(80),
     1IYS(80)
      READ(8,102)ICOM1,NC,ICOM2,BGR
      READ(8,103) (IXC(J),IYS(J),TA(J),J=1,NC)
      DO 20 J=1,NC
   20 IYS(J)=IYS(J) -BGR*TA(J)
C
C     FIND FIRST PRIMARY DISTANCE POINT
C
      DO 21 I=1,NP
   21 IX(I)=X(I)*10.0+.001
      IS=DS*10.0 + .001
      DO 1 I=1,NP
      IF(IX(I).EQ.IS)GO TO 2
    1 CONTINUE
C
C     INITIALIZE INDICES
C
    2 K=I
      J=0
      I=0
C
C     SCAN CASCADE FOR DISTANCE POINT .GE. PRIMARY POINT
C
    3 I=I+1
      IC=IXC(I)
      IF(I.GT.NC)GO TO 10
      IF(IC-IS)3,30,4
```

```
C
C    WHEN DISTANCE ENDPOINTS EQUAL
C
   30 J=J+1
      IYC(J)=IYS(I)
      GO TO 6
C
C    WHEN NOT EQUAL DO LINEAR INTERPOLATION
C
    4 Y2=IYS(I)
      Y1=IYS(I-1)
      X2=IXC(I)
      X1=IXC(I-1)
    5 Q=IS
      S=(Y2-Y1)/(X2-X1)
      B=Y1-S*X1
      YT=S*Q+B
      J=J+1
      IYC(J)=YT
C
C    CHECK IF NEXT PRIMARY DIST. IS WITHIN CURRENT
C    CASCADE POINTS; IF SO,
C    USE SAME POINTS FOR INTERPOLATION.
C
    6 K=K+1
      IF(K.GT.NP)GO TO 10
      IS=IX(K)
      IF(IC-IS),3,30,5
C
C    FINISH OFF
C
   10 NC=J
      DO 11 I=1,NC
   11 YC(I)=IYC(I)
      WRITE(6,101)(IX(I),IYC(I),I=1,NC)
  101 FORMAT(5(1X,I3,I6))
  102 FORMAT(4A4,54X,I2/4A4,F6.3)
  103 FORMAT(5(I3,I6,F4.1,1X))
 2000 RETURN
      END
```

APPENDIX III

Invariance of ANDC Analysis under Time Integration

Measurement of experimental decay curves requires collection of the emitted light over a finite section of the beam. This effective time integration of the decay curve can cause a significant deviation in the shape of the measured decay curve from the theoretical shape, depending on the relative sizes of the averaging length and the lifetime components of the decay curve. Due to the nature of ANDC analysis, although the variables involved (ie. areas and differences in intensity) are affected by this integration, the parameters obtained from the analysis (ie. slope and intercept) are, in principle, not affected by the value of the integration length, as will be shown in the following paragraphs.

If the "true" decay curve is given by

$$I_i(t) = Ae^{-\alpha_1 t} + Be^{-\alpha_2 t} + \ldots , \qquad \text{(A3.1)}$$

then the measured curve when the integration length is δ will be

$$I(t) = \int_{t-\delta/2}^{t+\delta/2} I_i(t')dt' .$$

Substitution of A3.1 into this then yields

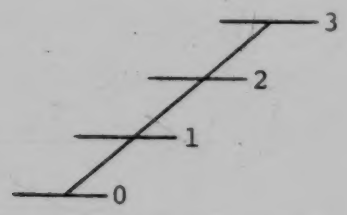
$$I(t) = E\left\{A\ \frac{sh(\alpha_1\delta/2)}{\alpha_1/2}\ e^{-\alpha_1 t} + B\ \frac{sh(\alpha_2\delta/2)}{\alpha_2/2}\ e^{-\alpha_2 t} + \ldots\right\}$$

$$= E\{AS_1e^{-\alpha_1 t} + BS_2e^{-\alpha_2 t} + \ldots\}\ ,\ \text{for } t \gg \delta/2 \text{ and where}$$

the common factor, δ, has been incleded in the detection efficiency, E, for the transition.

First, consider a four level decay scheme with only one direct cascade, as shown in Fig. A3.1 . ANDC theory states

Fig. A3.1 Four level decay scheme.



that $\tau_1 = a(I,F) - \xi b(I,F)$, where $\xi = \frac{E_{10}}{E_{21}} A_{10}\tau_1$,

$$a(I,F) = \frac{\int_I^F I_{10}(t)dt}{I_{10}(I)-I_{10}(F)} = \frac{P}{\Delta} \quad \text{and}$$

$$b(I,F) = \frac{\int_I^F I_{21}(t)dt}{I_{10}(I)-I_{10}(F)} = \frac{C}{\Delta}\ .$$

If a(I,F) is the "true" ANDC quantity, including efficiency, then for δ sufficiently large, the measured value of a(I,F) will be $a'(I,F) \neq a(I,F)$, and similarly for b(I,F) etc. In order for ANDC to remain valid, the condition $a(I,F)-b(I,F)\xi=a'-b'\xi$ must remain true for all (I,F). This

condition can be re-written, dropping the (I,F), as

$$\Delta'(P-\xi C) = \Delta(P'-\xi C') \quad . \tag{A3.2}$$

Expanding the quantities in this last expression into a sum of exponential terms yields

$$\begin{aligned}
P &= E_{10}\{P_1(e^{-\alpha_1 t_I}-e^{-\alpha_1 t_F})/\alpha_1 + P_2(e^{-\alpha_2 t}-e^{-\alpha_2 t})/\alpha_2 \\
&\qquad + P_3(e^{-\alpha_3 t}-e^{-\alpha_3 t})/\alpha_3\} \\
&= E_{10}\{P_1Z_1/\alpha_1 + P_2Z_2/\alpha_2 + P_3Z_3/\alpha_3\} \ , \\
C &= E_{21}\{C_2Z_2/\alpha_2 + C_3Z_3/\alpha_3\} \ , \\
\Delta &= E_{10}\{P_1Z_1 + P_2Z_2 + P_3Z_3\} \ , \\
P' &= E_{10}\{P_1S_1Z_1/\alpha_1 + P_2S_2Z_2/\alpha_2 + P_3S_3Z_3/\alpha_3\} \ , \\
C' &= E_{21}\{C_2S_2Z_2/\alpha_2 + C_3S_3Z_3/\alpha_3\} \ , \\
\Delta' &= E_{10}\{P_1S_1Z_1 + P_2S_2Z_2 + P_3S_3Z_3\} \ .
\end{aligned} \tag{A3.3}$$

The P_i and C_i may be expressed in terms of lifetimes, transition probabilities and initial level populations, as was discussed in Appendix 1. In order to remove the consideration of efficiency, one can write $\xi = qE_{10}/E_{21}$, where $q = A_{10}\tau_1$. The condition for the validity of ANDC analysis (eqn.A3.2) can now be written as

$$\begin{aligned}
&\{P_1S_1Z_1+P_2S_2Z_2+P_3S_3Z_3\}\{P_1Z_1/\alpha_1 + Z_2(P_2-qC_2)/\alpha_2 + \\
&\qquad + Z_3(P_3-qC_3)/\alpha_3\} = \\
&=\{P_1Z_1+P_2Z_2+P_3Z_3\}\{P_1S_1Z_1/\alpha_1 + S_2Z_2(P_2-qC_2)/\alpha_2 + S_3Z_3(P_3-qC_3)/\alpha_3\}
\end{aligned}$$

Note that the efficiency factors, E_{10} and E_{21}, have cancelled out.

This equation can be most easily verified in parts by equating the factors of each S_j.

$$S_2: \quad P_2Z_2\{P_1Z_1/\alpha_2 + Z_3\underline{(P_3-qC_3)}\} = (P_2-qC_2)Z_2\{P_1Z_1+P_3Z_3\}/\alpha_2$$
$$\alpha_3$$

It can be easily shown (see below) that in general

$$P_r-qC_r = P_r\alpha_r/\alpha_1 \ . \qquad \text{(A3.4)}$$

Therefore, the previous equation becomes

$$P_2\{P_1Z_1/\alpha_1 + \underline{P_3Z_3}\} = P_2\{P_1Z_1 + P_3Z_3\}/\alpha_1 \ ,$$
$$\alpha_1$$

or, $\qquad 1 = 1 \ .$

The other two factors give the same result, showing ANDC to be invariant under time integration in the four level case.

In order to prove its validity for any decay scheme, it first must be shown that

$$P_r-qC_r^d = P_r\alpha_r/\alpha_1 \quad , \text{ or,}$$

$$(1-qC_r^d/P_r) = \alpha_r/\alpha_1 \ .$$

Using the results of Appendix I, in general one can write

$$P_r=A_{10}\sum_{j=r}^{m}N_j(0)\left\{\frac{\prod_{i=1}^{j-1}A_{i+1,i}}{\prod_{\substack{k=1\\k\neq r}}^{m}(\alpha_k-\alpha_r)}\right\} \text{and} \quad C_r^d=A_{d1}\sum_{j=r}^{m}N_j(0)\left\{\frac{\prod_{i=d}^{j-1}A_{i+1,i}}{\prod_{\substack{k=d\\k\neq r}}^{m}(\alpha_k-\alpha_r)}\right\} \ .$$

Therefore,

$$C_r/P_r = A_{d1}/A_{10}\,\frac{A_{d1}}{(\alpha_1-\alpha_r)} = \frac{\alpha_1-\alpha_r}{A_{10}}\ , \quad \text{and}$$

$$1 - qC_r/P_r = 1 - \frac{A_{10}}{\alpha_1}\frac{C_r}{P_r} = \alpha_r/\alpha_1\ , \text{ proving eqn. A3.4 .}$$

In the case of the general decay scheme, eqn. A3.2 becomes (note that $C_1=0$),

$$\{\sum_i P_i S_i Z_i\}\{\sum_i Z_i \frac{(P_i - qC_i)}{\alpha_i}\} = \{\sum_i P_i Z_i\}\{\sum_i S_i Z_i (P_i - qC_r)/\alpha_i\} .$$

Substitution of eqn. A3.4 yields,

$$\{\sum_i P_i S_i Z_i\}\{\sum_i Z_i P_i/\alpha_i\} = \{\sum_i P_i Z_i\}\{\sum_i S_i Z_i P_i/\alpha_i\} .$$

Q.E.D.